

## PATENT COOPERATION TREATY

PCT

## NOTIFICATION OF ELECTION

(PCT Rule 61.2)

From the INTERNATIONAL BUREAU

To:

United States Patent and Trademark  
Office  
(Box PCT)  
Crystal Plaza 2  
Washington, DC 20231  
ÉTATS-UNIS D'AMÉRIQUE

in its capacity as elected Office

Date of mailing (day/month/year) 03 March 1999 (03.03.99)	
International application No. PCT/NL95/00153	Applicant's or agent's file reference X Sch/EN/2
International filing date (day/month/year) 27 April 1995 (27.04.95)	Priority date (day/month/year)
Applicant VAN DIJK, Dirk et al	

1. The designated Office is hereby notified of its election made:

☒ in the demand filed with the International Preliminary Examining Authority on:  
21 October 1996 (21.10.96)

☐ in a notice effecting later election filed with the International Bureau on:  
\_\_\_\_\_

2. The election ☒ was  
☐ was not

made before the expiration of 19 months from the priority date or, where Rule 32 applies, within the time limit under Rule 32.2(b).

The International Bureau of WIPO 34, chemin des Colombettes 1211 Geneva 20, Switzerland	Authorized officer F. Gateau
Facsimile No.: (41-22) 740.14.35	Telephone No.: (41-22) 338.83.38

Form PCT/IB/331 (July 1992)

2503428

## PATENT COOPERATION TREATY

## PCT

EL169834069

## INTERNATIONAL SEARCH REPORT

(PCT Article 18 and Rules 43 and 44)

Applicant's or agent's file reference <b>X Sch/EN/2</b>	<b>FOR FURTHER ACTION</b> see Notification of Transmittal of International Search Report (Form PCT/ISA/220) as well as, where applicable, item 5 below.	
International application No. <b>PCT/NL95/00153</b>	International filing date (day/month/year) <b>27/04/95</b>	(Earliest) Priority Date (day/month/year)
Applicant <b>AVIPLAST B.V. et al.</b>		

This international search report has been prepared by this International Searching Authority and is transmitted to the applicant according to Article 18. A copy is being transmitted to the International Bureau.

This international search report consists of a total of 5 sheets.

☒ It is also accompanied by a copy of each prior art document cited in this report.

1. ☐ Certain claims were found unsearchable (see Box I).
2. ☒ Unity of invention is lacking (see Box II).
3. ☐ The international application contains disclosure of a nucleotide and/or amino acid sequence listing and the international search was carried out on the basis of the sequence listing
  - ☐ filed with the international application.
  - ☐ furnished by the applicant separately from the international application,
    - ☐ but not accompanied by a statement to the effect that it did not include matter going beyond the disclosure in the international application as filed.
  - ☐ Transcribed by this Authority
4. With regard to the title,
  - ☒ the text is approved as submitted by the applicant.
  - ☐ the text has been established by this Authority to read as follows:
5. With regard to the abstract,
  - ☒ the text is approved as submitted by the applicant.
  - ☐ the text has been established, according to Rule 38.2(b), by this Authority as it appears in Box III. The applicant may, within one month from the date of mailing of this international search report, submit comments to this Authority.
6. The figure of the drawings to be published with the abstract is:  
Figure No. \_\_\_\_\_
  - ☐ as suggested by the applicant.
  - ☐ because the applicant failed to suggest a figure.
  - ☐ because this figure better characterizes the invention.

☒ None of the figures.

# INTERNATIONAL SEARCH REPORT

International application No.

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## Box I Observations where certain claims were found unsearchable (Continuation of item 1 of first sheet)

This international search report has not been established in respect of certain claims under Article 17(2)(a) for the following reasons:

1. ☐ Claims Nos.:  
because they relate to subject matter not required to be searched by this Authority, namely:
2. ☐ Claims Nos.:  
because they relate to parts of the international application that do not comply with the prescribed requirements to such an extent that no meaningful international search can be carried out, specifically:
3. ☐ Claims Nos.:  
because they are dependent claims and are not drafted in accordance with the second and third sentences of Rule 6.4(a).

## Box II Observations where unity of invention is lacking (Continuation of item 2 of first sheet)

This International Searching Authority found multiple inventions in this international application, as follows:

- 1.- claims 1-19: Plastic composite, products and process
- 2.- claims 20-27: Apparatus and its technical features
- 3.- claims 28-29: Method, using after-treatment
- 3.- claim 30: method, starting from prepolymer

1. ☒ As all required additional search fees were timely paid by the applicant, this international search report covers all searchable claims:
2. ☐ As all searchable claims could be searched without effort justifying an additional fee, this Authority did not invite payment of any additional fee.
3. ☐ As only some of the required additional search fees were timely paid by the applicant, this international search report covers only those claims for which fees were paid, specifically claims Nos.:
4. ☐ No required additional search fees were timely paid by the applicant. Consequently, this international search report is restricted to the invention first mentioned in the claims; it is covered by claims Nos.:

Remark on Protest

- ☒ The additional search fees were accompanied by the applicant's protest.
- ☐ No protest accompanied the payment of additional search fees.

## INTERNATIONAL SEARCH REPORT

International Application No

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A. CLASSIFICATION OF SUBJECT MATTER  
IPC 6 C08K7/00 C08J5/04 B29C47/00 B32B27/04 C08L101/00  
B29C71/02 C08F2/44 B29C67/24

According to International Patent Classification (IPC) or to both national classification and IPC

## B. FIELDS SEARCHED

Minimum documentation searched (classification system followed by classification symbols)

IPC 6 C08K B29C

Documentation searched other than minimum documentation to the extent that such documents are included in the fields searched

Electronic data base consulted during the international search (name of data base and, where practical, search terms used)

## C. DOCUMENTS CONSIDERED TO BE RELEVANT

Category *	Citation of document, with indication, where appropriate, of the relevant passages	Relevant to claim No.
A	EP,A,0 319 589 (NAMBA) 14 June 1989	1,6,12
A	see page 7, line 1 - line 16; claims 1-12	1-13
A	EP,A,0 591 801 (UNITIKA LTD.) 13 April 1994	1-19
A	see page 3, line 31 - page 4, line 17	1,5
A	see claims 1,10,11,14	1-13
A	US,A,5 082 605 (J.G. BROOKS ET AL.) 21 January 1992	
	cited in the application	
	see column 4, line 21 - line 35	
	see column 7, line 7 - line 24; claims 1-7,10-15	
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☒ Further documents are listed in the continuation of box C.☒ Patent family members are listed in annex.

## \* Special categories of cited documents :

- \*A\* document defining the general state of the art which is not considered to be of particular relevance
- \*E\* earlier document but published on or after the international filing date
- \*L\* document which may throw doubts on priority claim(s) or which is cited to establish the publication date of another citation or other special reason (as specified)
- \*O\* document referring to an oral disclosure, use, exhibition or other means
- \*P\* document published prior to the international filing date but later than the priority date claimed

\*T\* later document published after the international filing date or priority date and not in conflict with the application but cited to understand the principle or theory underlying the invention

\*X\* document of particular relevance; the claimed invention cannot be considered novel or cannot be considered to involve an inventive step when the document is taken alone

\*Y\* document of particular relevance; the claimed invention cannot be considered to involve an inventive step when the document is combined with one or more other such documents, such combination being obvious to a person skilled in the art.

\*&\* document member of the same patent family

Date of the actual completion of the international search

29 March 1996

Date of mailing of the international search report

Name and mailing address of the ISA

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Authorized officer

Engel, S

# INTERNATIONAL SEARCH REPORT

International Application No  
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## C.(Continuation) DOCUMENTS CONSIDERED TO BE RELEVANT

Category *	Citation of document, with indication, where appropriate, of the relevant passages	Relevant to claim No.
A	DE,A,28 11 795 (POLYPLASTICS) 12 October 1978 see page 5, paragraph 2 see page 7, paragraph 2; claims 1-4; example 1 ---	1,5
A	DATABASE WPI Section Ch, Week 8504 Derwent Publications Ltd., London, GB; Class A17, AN 85-024748 & SU,A,1 100 288 ( ZLOBINA V A ) , 30 June 1984 see abstract ---	1-13
A	DATABASE WPI Section Ch, Week 8504 Derwent Publications Ltd., London, GB; Class A17, AN 85-022209 & JP,A,59 217 743 ( KASAI KOGYO KK ) , 7 December 1984 see abstract ---	1-13
A	PATENT ABSTRACTS OF JAPAN vol. 94 no. 00010 & JP,A,62 079615 (CHISSO CORP) 4 October 1994, see abstract & DATABASE WPI Week 9444 Derwent Publications Ltd., London, GB; AN 94-354843 & JP,A,62 079 615 (CHISSO) , 4 October 1994 see abstract ---	1-19
E	NL,A,9 302 125 (AVIPLAST) 3 July 1995 see page 4, line 30 - page 5, line 2 see page 5, line 11 - line 28 see figures 1,2 see claims 1,11 & NL,A,9 400 308 (AVIPLAST)  see page 3, line 23 - line 39 see page 5, line 13 - page 6, line 20; claims 1,4,5,8,10,11; figure 1 ---	20
E		1,12,20, 21
A	US,A,5 096 406 (BROOKS ET AL.) 17 March 1992 see figures & US,A,5 082 605 (BROOKS ET AL.) cited in the application see column 7, paragraph 2 ---	20,21
A		20

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# INTERNATIONAL SEARCH REPORT

International Application No  
PCT/NL 95/00153

## C.(Continuation) DOCUMENTS CONSIDERED TO BE RELEVANT

Category *	Citation of document, with indication, where appropriate, of the relevant passages	Relevant to claim No.
A	PATENT ABSTRACTS OF JAPAN vol. 014 no. 305 (M-0992) ,29 June 1990 & JP,A,02 098429 (NHK SPRING CO LTD) 10 April 1990, see abstract ---	1,28,29
A	EP,A,0 008 143 (SOLVAY & CIE.) 20 February 1980 see page 3, line 33 - page 4, line 5 see page 6, line 9 - line 11 see page 8, line 8 - line 15 ---	1,28,29
A	US,A,5 401 154 (M.M. SARGENT) 28 March 1995 see column 3, line 19 - line 28 ---	1,28,29
A	DATABASE WPI Section Ch, Week 8522 Derwent Publications Ltd., London, GB; Class A32, AN 85-131721 & JP,A,60 069 161 ( TOSHIBA KK) , 19 April 1985 see abstract -----	1,30

# INTERNATIONAL SEARCH REPORT

Information on patent family members

International Application No

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Patent document cited in search report	Publication date	Patent family member(s)	Publication date
EP-A-319589	14-06-89	JP-A- 1004652 DE-D- 3854646 DE-T- 3854646 AT-T- 129732 AU-B- 605608 CA-A- 1318067 WO-A- 8810286	09-01-89 07-12-95 21-03-96 15-11-95 17-01-91 18-05-93 29-12-88
EP-A-591801	13-04-94	JP-A- 6115040	26-04-94
US-A-5082605	21-01-92	US-A- 5088910 US-A- 5096046 US-A- 5096406 US-A- 5213021	18-02-92 17-03-92 17-03-92 25-05-93
DE-A-2811795	12-10-78	JP-C- 1513968 JP-A- 53121843 JP-B- 62029450 AU-B- 522577 AU-B- 3465878 BE-A- 865101 FR-A,B 2393016 GB-A- 1587894 NL-A- 7803537	24-08-89 24-10-78 26-06-87 17-06-82 04-10-79 17-07-78 29-12-78 15-04-81 03-10-78
NL-A-9302125	03-07-95	NL-A- 9400308	03-07-95
US-A-5096406	17-03-92	US-A- 5088910 US-A- 5096046 US-A- 5082605 US-A- 5213021	18-02-92 17-03-92 21-01-92 25-05-93
EP-A-08143	20-02-80	FR-A- 2433031 AT-T- 1155 CA-A- 1146325 JP-A- 55025495 US-A- 4380522	07-03-80 15-06-82 17-05-83 23-02-80 19-04-83
US-A-5401154	28-03-95	NONE	

## PATENT COOPERATION TREATY

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## INTERNATIONAL PRELIMINARY EXAMINATION REPORT

(PCT Article 36 and Rule 70)

Applicant's or agent's file reference <b>95 6171 PC</b>	FOR FURTHER ACTION See Notification of Transmittal of International Preliminary Examination Report (Form PCT/IPEA/416)	
International application No. <b>PCT/NL 95/ 00153</b>	International filing date (day/month/year) <b>27/04/1995</b>	Priority date (day/month/year) <b>[27/04/1995] NONE</b>
International Patent Classification (IPC) or national classification and IPC <b>C08K7/00</b>		
Applicant <b>AVIPLAST B.V. et al.</b>		

1. This international preliminary examination report has been prepared by this International Preliminary Examining Authority and is transmitted to the applicant according to Article 36.



2. This REPORT consists of a total of 7 sheets, including this cover sheet.

☒ This report is also accompanied by ANNEXES, i.e., sheets of the description, claims and/or drawings which have been amended and are the basis for this report and/or sheets containing rectifications made before this Authority (see Rule 70.16 and Section 607 of the Administrative Instructions under the PCT).

These annexes consists of a total of 9 sheets.

3. This report contains indications and corresponding pages relating to the following items:

- I ☒ Basis of the report
- II ☐ Priority
- III ☐ Non-establishment of opinion with regard to novelty, inventive step and industrial applicability
- IV ☐ Lack of unity of invention
- V ☒ Reasoned statement under Article 35(2) with regard to novelty, inventive step or industrial applicability; citations and explanations supporting such statement
- VI ☐ Certain documents cited
- VII ☒ Certain defects in the international application
- VIII ☒ Certain observations on the international application

Date of submission of the demand <b>21/10/1996</b>	Date of completion of this report <b>12.08.97</b>
Name and mailing address of the IPEA/  European Patent Office D-80298 Munich Tel. (+49-89) 2399-0, Tx: 523656 epmu d Fax: (+49-89) 2399-4465	Authorized officer  <b>F. Hein</b> Telephone No. <b>-2134</b>



## INTERNATIONAL PRELIMINARY EXAMINATION REPORT

## I. Basis of the report

1. This report has been drawn up on the basis of (Replacement sheets which have been furnished to the receiving Office in response to an invitation under Article 14 are referred to in this report as "originally filed" and are not annexed to the report since they do not contain amendments.):

☐ the international application as originally filed.

☒ the description, pages 3-10, 12-17 \_\_\_\_\_, as originally filed,  
pages \_\_\_\_\_, filed with the demand,  
pages 1, 1B, 1C, 2, 11 \_\_\_\_\_, filed with the letter of 13/06/97,  
pages 1A \_\_\_\_\_, filed with the letter of 24/06/97,

☒ the claims, Nos. \_\_\_\_\_, as originally filed,  
Nos. \_\_\_\_\_, as amended under Article 19,  
Nos. \_\_\_\_\_, filed with the demand,  
Nos. 1-7 \_\_\_\_\_, filed with the letter of 24/06/97,  
Nos. \_\_\_\_\_, filed with the letter of \_\_\_\_\_,

☒ the drawings, sheets/fig 1/3 - 3/3 \_\_\_\_\_, as originally filed,  
sheets/fig \_\_\_\_\_, filed with the demand,  
sheets/fig \_\_\_\_\_, filed with the letter of \_\_\_\_\_,  
sheets/fig \_\_\_\_\_, filed with the letter of \_\_\_\_\_.

2. The amendments have resulted in the cancellation of:

☐ the description, pages \_\_\_\_\_.  
☐ the claims, Nos. \_\_\_\_\_.  
☐ the drawings, sheets/fig \_\_\_\_\_.

3. ☒ ~~This report has been established as if (some of) the amendments had not been made, since they have been considered to go beyond the disclosure as filed (Rule 70.2(c)).~~

The amendments are considered to go beyond the disclosure as filed [Rule 70.2(c) PCT]:

1. It is unclear which passages of the application as filed support the following features of operative Claim 1:

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- (a) "elongated (particles)";
- (b) "subsequently at least a part of the particles is oriented to a flow direction";
- (c) "with a passage opening which connects onto the outlets of the orienting means";
- (d) "wherein the form of the passage opening of the mould head (corresponds with the desired cross sectional form of the product)";
- (e) "(the larger particles) ... are oriented to a dominant orientation".

2.1. It is clear from the description on p. 5, l. 14-35, p. 6, l. 28-38, p. 7, l. 1-22, original Claim 20 and Fig. 1 and 3 that the following features are essential to the performance of the "compushtruder":

- (1) a compounder operating at low pressure;
- (2) a rotating displacement pump;
- (3) a distribution head;

Generalized apparatus claims (2-7) which are devoid of said essential features therefore fail to meet the requirements of Rule 70.2 (c) PCT.

2.2. It is unclear which passages of the application as filed support the following features of operative Claim 2:

- (a) "(orienting means) for orienting at least a part the elongated particles substantially into the flow direction";
- (b) "with a passage opening which connects onto the outlets of the orienting means";
- (c) "wherein the form of the passage opening of the mould head (corresponds with the desired cross sectional form of the product)";
- (d) "(temperature curve) in the flow direction in the passage opening of the mould head".

- 
3. It is unclear which passages of the application as filed support the following features of operative Claim 4:
- (a) "(dimensioned relative to) the elongated particles";
  - (b) "... passage of the particles) longer than a certain predetermined length in a direction which corresponds other than to a dominating extent with the flow direction".
4. It is unclear which passage of the application as filed supports the following features of operative Claim 5:  
"shortest particles", longest particles".

To summarize, the operative set of claims is not considered to meet the requirements of Rule 70.2 (c) PCT.

4. Additional observations, if necessary:

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V. Reasoned statement under Article 35(2) with regard to novelty, inventive step and industrial applicability; citations and explanations supporting such statement

1. STATEMENT

Novelty (N)	Claims 1-7 _____	YES
	Claims _____	NO
Inventive Step (IS)	Claims 1-7 _____	YES
	Claims _____	NO
Industrial Applicability (IA)	Claims 1-7 _____	YES
	Claims _____	NO

2. CITATIONS AND EXPLANATIONS

The prior art cited in the International Search Report is silent on a method or an apparatus for manufacturing reinforced plastic-based composite products wherein a prismatic mould head having a specific temperature profile is employed.

Hence, claims pertaining to said method and apparatus which are in accordance with Rule 70.2(c) PCT are considered to meet the requirements of Art. 33 (2)-(4) PCT (cf. original claims 20-28).

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VII. Certain defects in the international application

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The following defects in the form or contents of the international application have been noted:

1. Although Claims 1 and 2 are drafted in the two-part form the feature "prismatic mould head" is incorrectly placed in the preamble, as it is not disclosed in US-A-5 082 605 (D2) or US-A-5 096 046 (Rule 6.3 (b) PCT).
2. In the first part of the example only ligneous fibrous particles having a length of 0.6 - 3 mm are employed. It is not clear how fibrous particles having a length of 1.9 mm adopt a random orientation when the fibrous particles having a length of 2 - 3 mm are "oriented to a dominant orientation" (i.e. > 50 %).

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VIII. Certain observations on the international application

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The following observations on the clarity of the claims, description, and drawings or on the question whether the claims are fully supported by the description, are made:

The meaning of features 1(b), 1(e) and 3(b) enumerated under point I.3. above is unclear (Art. 6 PCT).

Method and apparatus for manufacturing a plastic based composite product

The invention relates to a method for manufacturing a plastic-based composite product which consists at least partially of a plastic in which a material consisting substantially of elongated particles is homogeneously embedded, wherein first the particles are mixed with the plastic and plasticized and subsequently at least a part of the particles is oriented, and wherein subsequently the mixture of the plastic and the particles passes a substantially prismatic mould head with a passage opening which connects onto the outlets of the orienting means, wherein the form of the passage opening of the mould head corresponds with the desired cross sectional form of the product. The invention also relates to an apparatus for carrying out such a method.

15

Such a method is known from the US patent US-A 5,082,605. This known method is particularly meant for producing a composite comprising wood particles which are embedded in a polymeric matrix. However, the properties of the composite are inferior in comparison with, for example, natural wood, since the polymeric matrix dominates the properties of the composite. The wood particles are relatively large and function as a filler. Therefor these wood particles hardly influence the mechanical properties, such as strength. The method as described in this US patent is an extrusion method. After leaving the extruder die, rollers are used to maintain dimensional stability of the extrudate. These rollers are adapted to exert only minimal pressure on the extrudate. This minimal pressure can only be exerted where the rollers are in contact with the extrudate. As a consequence the pressure on the composite material is very low and not uniform. With such extrusion methods an afterflow in the calibration unit is normally necessary to enable replenishing of the material deficit resulting from shrinkage. This after-flow causes delamination and a decrease in cohesion between the particles and the plastic. This results in mechanical properties with relatively low

values. Therefor, the application of the products which are made according to this known process are limited to ornamentation, covering, fence palings, etc.

5 The US patent US 5,096,406 describes an extruder assembly for composite materials, in particular materials comprising elongated wood particles which are embedded in a polymeric material. The composite material is extruded under high pressure, which can cause breaking of the particles. After  
10 the composite material leaves the mould head, the pressure on the material falls, which leads to the same drawback as the method according to the US patent 5,082,605.

The object of the invention is a method and an apparatus to  
15 produce a composite product at low cost but which meets high standards for mechanical properties comparable to the properties of for example wood and also meets high standards for for instance fire resistance, weather resistance, non-aging properties and the like, wherein a large amount  
20 of particles can be applied without the risk of delaminations and the like.

This object of the invention is achieved with a method wherein the particles have a length between 0.2 and about 6  
25 mm wherein the larger particles, which have a length of about 2 - 6 mm, are oriented to a dominant orientation while the smaller particles show a random orientation, and in that after entering the mould head the finished product has a temperature above the Vicat softening temperature of  
30 the applied plastic respectively the highest Vicat softening temperature of all the applied plastics, modelling the product to a desired shape, and cooling the product in this shape to below the said Vicat softening temperature while maintaining a chosen pressure. With this measure, a con-  
35 solidation of the composite material is achieved which prevents delamination and matrix failure. As a result, a composite material is obtained with mechanical properties which meet high standards.



The object of the invention is also achieved with an apparatus of the above mentioned type, wherein the length of the mould head and the temperature curve in the flow direction in the passage opening of the mould head are such that at the end of the passage opening the product has cooled to below its Vicat softening temperature. With such a apparatus it has become possible to cool the composite material in a controllable way under a certain pressure until below the Vicat softening temperature within a continuous process.

With the method and the apparatus according to the invention, a composite material comprising particles which are embedded in a polymeric matrix, can be manufactured without waste of particles and, if desired, in a continuous process, which product does not only possess mechanical and physical/chemical properties corresponding with wood but can also be manufactured on the basis wood, for instance inferior wood, which can be classified as interior wood, exterior wood or construction wood.

The particles may be fibers or may be plates of wood or, for instance, mica.

Such composite material, which is manufactured according to the invention, will hereinafter be designated as technical wood. The obtained product can be applied at locations where tropical hardwood is still indicated.

Preferably, the plastic is a thermoplastic polymer material, in particular at least one polyolefin or one polymer on a basis of styrene, wherein

a. the transverse dimension of the larger wood particles is preferably such that the ratio of the length in the fiber direction of the particles to this transverse dimension amounts to a minimum of 4, but preferably lies in the range of 6 - 80;

b. The wood particles are present in a quantity of 40 - 80 % by mass, but preferably from 50 - 70 % by mass in

relation to the mass of product;

c. The obtained product complies minimally with the following requirements relating to mechanical properties in

- . bending strength in the fiber direction: 8 MPa
- 5 . bending modulus in the fiber direction: 3 GPa
- . tensile strength in the fiber direction: 6 MPa
- . tensile stress modulus in the fiber direction: 3 GPa
- . tensile strength transversely of fiber direction: 0.3 MPa
- 10 . tensile strength modulus transversely of fiber direction: 1 GPa

*continue page 2, line 35 of original text*

## CLAIMS

1. Method for manufacturing a plastic-based composite product which consists at least partially of a plastic in which a material consisting substantially of elongated particles is homogeneously embedded, wherein first the  
5 particles are mixed with the plastic and plasticized and subsequently at least a part of the particles is oriented to a flow direction, and wherein subsequently the mixture of the plastic and the particles passes a substantially prismatic mould head with a passage opening which connects  
10 onto the outlets of the orienting means, wherein the form of the passage opening of the mould head corresponds with the desired cross sectional form of the product **characterized in that**, the particles have a length between 0.2 and about 6 mm wherein the larger particles, which have a  
15 length of about 2 - 6 mm, are oriented to a dominant orientation while the smaller particles show a random orientation, and in that after entering the mould head the product has a temperature above the Vicat softening temperature of the applied plastic respectively the highest Vicat softening temperature of all the applied plastics, modelling the  
20 product to a desired shape, and cooling the product in this shape to below the said Vicat softening temperature while maintaining a chosen pressure.

25 2. Apparatus for manufacturing a plastic based composite product, which product consists at least partially of a plastic in which elongated particles are homogeneously embedded, which apparatus comprises:

- means for plasticizing a mixture consisting substantially of the plastic and the particles, which particles  
30 are mixed with the plastic either beforehand or in the compounder, and pressing the plastic mixture to the outside via an outlet;
- orienting means for orienting at least a part the  
35 elongated particles substantially into the flow direction;
- a substantially prismatic mould head with a passage

opening which connects onto the outlets of the orienting means, wherein the form of the passage opening of the mould head corresponds with the desired cross sectional form of the product **characterized in that**, the length of the mould head and the temperature curve in the flow direction in the passage opening of the mould head are such that at the end of the passage opening the product has cooled to below its Vicat softening temperature.

10 3. Apparatus according to claim 2 **characterized in that** the plasticizing means consist of a compounder operating at low pressure and in that the apparatus further comprises a rotating displacement pump for further transporting the plastic mixture.

15 4. Apparatus as claimed in claim 2 or 3 **characterized in that**, the apparatus comprises a distribution head further guiding the mixture in order to further transport the plastic mixture substantially as a plug flow, wherein the orienting means comprise at least one bundle of substantially parallel channels through which the plastic mixture can flow, which channels are dimensioned relative to the elongated particles such that they are too small to allow passage of the particles longer than a certain predetermined length in a direction which corresponds other than to a dominating extent with the flow direction.

5. Apparatus as claimed in one of the preceding claims 2 - 4 **characterized in that**, the linear transverse dimensions of at least the parts of the channels located furthest downstream amount to a minimum of 2x the length of the shortest particles and a maximum of 1.5x the length of the longest particles.

35 6. Apparatus as claimed in one of the preceding claims 2 - 5 **characterized in that**, the orienting means comprises at least two bundles of channels mutually connecting in series, wherein the transverse dimensions of the channels of a bundle located further downstream are smaller than

those of the channels of a bundle located further upstream.

7. Apparatus as claimed in one of the preceding claims 2  
5 - 6 **characterized in that**, at least one feed for adding  
particle material and/or additives to the plastic connects  
to the compounder.

405 Rec'd PCT/PTO 27 OCT 1998

Method and apparatus for manufacturing a plastic based composite product

Not entered

The invention relates to a method for manufacturing a plastic-based composite product which consists at least partially of a plastic in which a material consisting substantially of elongated particles is homogeneously embedded, wherein first the particles are mixed with the plastic and plasticized and subsequently at least a part of the particles is oriented, and wherein subsequently the mixture of the plastic and the particles passes a substantially prismatic mould head with a passage opening which connects onto the outlets of the orienting means, wherein the form of the passage opening of the mould head corresponds with the desired cross sectional form of the product. The invention also relates to an apparatus for carrying out such a method.

15

Such a method is known from the US patent US-A 5,082,605. This known method is particularly meant for producing a composite comprising wood particles which are embedded in a polymeric matrix. However, the properties of the composite are inferior in comparison with, for example, natural wood, since the polymeric matrix dominates the properties of the composite. The wood particles are relatively large and function as a filler. Therefor these wood particles hardly influence the mechanical properties, such as strength. The method as described in this US patent is an extrusion method. After leaving the extruder die, rollers are used to maintain dimensional stability of the extrudate. These rollers are adapted to exert only minimal pressure on the extrudate. This minimal pressure can only be exerted where the rollers are in contact with the extrudate. As a consequence the pressure on the composite material is very low and not uniform. With such extrusion methods an afterflow in the calibration unit is normally necessary to enable replenishing of the material deficit resulting from shrinkage. This after-flow causes delamination and a decrease in cohesion between the particles and the plastic. This results in mechanical properties with relatively low

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values. Therefor, the application of the products which are made according to this known process are limited to ornamentation, covering, fence palings, etc.

- 5 The US patent US 5,096,406 describes an extruder assembly for composite materials, in particular materials comprising elongated wood particles which are embedded in a polymeric material. The composite material is extruded under high pressure, which can cause breaking of the particles. After  
10 the composite material leaves the mould head, the pressure on the material falls, which leads to the same drawback as the method according to the US patent 5,082,605.

The object of the invention is a method and an apparatus to  
15 produce a composite product at low cost but which meets high standards for mechanical properties comparable to the properties of for example wood and also meets high standards for for instance fire resistance, weather resistance, non-aging properties and the like, wherein a large amount  
20 of particles can be applied without the risk of delaminations and the like.

This object of the invention is achieved with a method wherein the particles have a length between 0.2 and about 6  
25 mm wherein the larger particles, which have a length of about 2 - 6 mm, are oriented to a dominant orientation while the smaller particles show a random orientation, and in that after entering the mould head the finished product is heated to a temperature above the Vicat softening temperature of the applied plastic respectively the highest  
30 Vicat softening temperature of all the applied plastics, modelling the product to a desired shape, and cooling the product in this shape to below the said Vicat softening temperature while maintaining a chosen pressure. With this  
35 measure, a consolidation of the composite material is achieved which prevents delamination and matrix failure. As a result, a composite material is obtained with mechanical properties which meet high standards.

The object of the invention is also achieved with an apparatus of the above mentioned type, wherein the length of the mould head and the temperature curve in the flow direction in the passage opening of the mould head are such that at the end of the passage opening the product has cooled to below its Vicat softening temperature. With such a apparatus it has become possible to cool the composite material in a controllable way under a certain pressure until below the Vicat softening temperature within a continuous process.

With the method and the apparatus according to the invention, a composite material comprising particles which are embedded in a polymeric matrix, can be manufactured without waste of particles and, if desired, in a continuous process, which product does not only possess mechanical and physical/chemical properties corresponding with wood but can also be manufactured on the basis wood, for instance inferior wood, which can be classified as interior wood, exterior wood or construction wood.

The particles may be fibers or may be plates of wood or, for instance, mica.

Such composite material, which is manufactured according to the invention, will hereinafter be designated as technical wood. The obtained product can be applied at locations where tropical hardwood is still indicated.

Preferably, the plastic is a thermoplastic polymer material, in particular at least one polyolefin or one polymer on a basis of styrene, wherein

a. the transverse dimension of the larger wood particles is preferably such that the ratio of the length in the fiber direction of the particles to this transverse dimension amounts to a minimum of 4, but preferably lies in the range of 6 - 80;

b. The wood particles are present in a quantity of 40 - 80 % by mass, but preferably from 50 - 70 % by mass in



relation to the mass of product;

- c. The obtained product complies minimally with the following requirements relating to mechanical properties in
- . bending strength in the fiber direction: 8 MPa
  - 5 . bending modulus in the fiber direction: 3 GPa
  - . tensile strength in the fiber direction: 6 MPa
  - . tensile stress modulus in the fiber direction: 3 GPa
  - . tensile strength transversely of fiber direction: 0.3 MPa
  - 10 . tensile strength modulus transversely of fiber direction: 1 GPa

*continue page 2, line 35 of original text*

~~/material and the plastic is a thermoplastic polymer material~~  
 in particular at least one polyolefin or one polymer on a  
 basis of styrene, wherein

- a. the transverse dimension of the large wood particles is  
 5 preferably such that the ratio of the length in the principal  
 direction of the particles to this transverse dimension  
 amounts to a minimum of 4, but preferably lies in the range  
 of 6-80.
- b. the wood particles are present in a quantity of 40-80% by  
 10 mass, but preferably from 50 to 70% by mass in relation to  
 the mass of product.
- c. the obtained product complies minimally with the following  
 requirements relating to mechanical properties in
  - . bending strength in the fibre direction: 8 MPa
  - 15 . bending modulus in the fibre direction: 3 GPa
  - . tensile strength in the fibre direction: 6 MPa
  - . tensile stress modulus in fibre direction: 3 GPa
  - . tensile strength transversely of fibre direction: 0.3 MPa
  - . tensile stress modulus transversely of fibre direction:  
 20 1GPa.

The invention herewith provides a product which in the  
 manner to be described hereinbelow can be manufactured on a  
 basis of wood without waste and, if desired, as a continuous  
 25 process, which product not only possesses mechanical and  
 physical/chemical properties corresponding with wood but which  
 product can be manufactured on the basis of basic wood which  
 can if desired be inferior and can be classified as interior  
 wood, exterior wood or construction wood. This product  
 30 according to the invention will hereinafter be designated as  
 technical wood.

The invention provides the possibility of applying the  
 obtained product at locations where tropical hardwood is still  
 indicated.

35 For this purpose particles with tensile strength such as  
 wood particles, preferably originating from waste wood or wood  
 waste, optionally together with suitable strengthening  
 particles, are predominantly oriented and bound by means of a  
 thermoplastic polymer, preferably on a basis of olefins or  
 40 styrene, wherein the proportion of applied particles is extreme

Bending tests (ISO 178):

Maximum bending load	32.0 MPa	0.3 MPa
Bending modulus	3.849 GPa	0.098 GPa

5

Addition of 10% glass or flax fibre with a length of 4 mm and L/D 150 to 400 gives values about 25% higher than shown above.

10 A product can in principle be manufactured in any suitable manner, starting from pre-compounded material or semi-manufacture, for instance by (isostatic) pressing, injection moulding, extruding, compushtruding.

15 A specific embodiment has the feature that the product consists substantially of a laminate comprising a plastic-based composite layer according to the above stated specifications; a first skin layer adhered thereto on one side and possessing chosen properties; optionally a second skin layer adhered thereto on the other side and possessing chosen properties; and which layers are  
20 mutually adhered by for instance glueing, welding, mirror-welding, with an infrared laser, with hot air or other suitable treatment.

Another embodiment is characterized by at least one additive for obtaining desired properties, which additive  
25 is added to the process flow at a suitable position in the compushtruding process during manufacture of the product.

The said chosen properties of the skin layers may relate for instance to the adherability of paint, lacquer  
30 and glue.

A variant of this latter embodiment has the feature that the skin layer or at least one of the skin layers is of the type according to the invention and the long particle orientation thereof has a chosen direction  
35 relative to this orientation of the first mentioned plastic layer. Specific mechanical properties can hereby be achieved. ~~These can be based for instance on the stringer effect~~

# PATENT COOPERATION TREATY

# PCT

# EL169834069



## INTERNATIONAL PRELIMINARY EXAMINATION REPORT

(PCT Article 36 and Rule 70)

Applicant's or agent's file reference <b>95 6171 PC</b>	<b>FOR FURTHER ACTION</b> See Notification of Transmittal of International Preliminary Examination Report (Form PCT/IPEA/416)	
International application No. <b>PCT/NL 95/ 00153</b>	International filing date (day/month/year) <b>27/04/1995</b>	Priority date (day/month/year) <b>27/04/1995</b>
International Patent Classification (IPC) or national classification and IPC <div style="text-align: center;"><b>C08K7/00</b></div>		
Applicant <b>AVIPLAST B.V. et al.</b>		

1. This international preliminary examination report has been prepared by this International Preliminary Examining Authority and is transmitted to the applicant according to Article 36.
2. This REPORT consists of a total of 16 sheets, including this cover sheet.  
☒ This report is also accompanied by ANNEXES, i.e., sheets of the description, claims and/or drawings which have been amended and are the basis for this report and/or sheets containing rectifications made before this Authority (see Rule 70.16 and Section 607 of the Administrative Instructions under the PCT).  
 These annexes consists of a total of 9 sheets.

3. This report contains indications and corresponding pages relating to the following items:
  - I ☒ Basis of the report
  - II ☐ Priority
  - III ☐ Non-establishment of opinion with regard to novelty, inventive step and industrial applicability
  - IV ☐ Lack of unity of invention
  - V ☒ Reasoned statement under Article 35(2) with regard to novelty, inventive step or industrial applicability; citations and explanations supporting such statement
  - VI ☐ Certain documents cited
  - VII ☒ Certain defects in the international application
  - VIII ☒ Certain observations on the international application

Date of submission of the demand <b>21/10/1996</b>	Date of completion of this report <b>12. 08. 97</b>
Name and mailing address of the IPEA/  <b>European Patent Office          D-80298 Munich          Tel. (+49-89) 2399-0, Tx: 523656 epmu d          Fax: (+49-89) 2399-4465</b>	Authorized officer <div style="text-align: center;">   <b>F. Hein</b> </div> Telephone No. <b>-2134</b>

INTERNATIONAL PRELIMINARY EXAMINATION REPORT

Intern. application No.

PCT/NL95/00153

I. Basis of the report

1. This report has been drawn up on the basis of (Replacement sheets which have been furnished to the receiving Office in response to an invitation under Article 14 are referred to in this report as "originally filed" and are not annexed to the report since they do not contain amendments.):

☐ the international application as originally filed.

☒ the description, pages 3-10, 12-17 \_\_\_\_\_, as originally filed,  
pages \_\_\_\_\_, filed with the demand,  
pages 1, 1B, 1C, 2, 11 \_\_\_\_\_, filed with the letter of 13/06/97,  
pages 1A \_\_\_\_\_, filed with the letter of 24/06/97,

☒ the claims, Nos. \_\_\_\_\_, as originally filed,  
Nos. \_\_\_\_\_, as amended under Article 19,  
Nos. \_\_\_\_\_, filed with the demand,  
Nos. 1-7 \_\_\_\_\_, filed with the letter of 24/06/97,  
Nos. \_\_\_\_\_, filed with the letter of \_\_\_\_\_,

☒ the drawings, sheets/fig 1/3 - 3/3 \_\_\_\_\_, as originally filed,  
sheets/fig \_\_\_\_\_, filed with the demand,  
sheets/fig \_\_\_\_\_, filed with the letter of \_\_\_\_\_,  
sheets/fig \_\_\_\_\_, filed with the letter of \_\_\_\_\_.

2. The amendments have resulted in the cancellation of:

☐ the description, pages \_\_\_\_\_.  
☐ the claims, Nos. \_\_\_\_\_.  
☐ the drawings, sheets/fig \_\_\_\_\_.

3. ☒ ~~This report has been established as if (some of) the amendments had not been made, since they have been considered to go beyond the disclosure as filed (Rule 70.2(c)).~~

The amendments are considered to go beyond the disclosure as filed [Rule 70.2(c) PCT]:

1. It is unclear which passages of the application as filed support the following features of operative Claim 1:

- (a) "elongated (particles)";
- (b) "subsequently at least a part of the particles is oriented to a flow direction";
- (c) "with a passage opening which connects onto the outlets of the orienting means";
- (d) "wherein the form of the passage opening of the mould head (corresponds with the desired cross sectional form of the product)";
- (e) "(the larger particles) ... are oriented to a dominant orientation".

2.1. It is clear from the description on p. 5, l. 14-35, p. 6, l. 28-38, p. 7, l. 1-22, original Claim 20 and Fig. 1 and 3 that the following features are essential to the performance of the "compushtruder":

- (1) a compounder operating at low pressure;
- (2) a rotating displacement pump;
- (3) a distribution head;

Generalized apparatus claims (2-7) which are devoid of said essential features therefore fail to meet the requirements of Rule 70.2 (c) PCT.

2.2. It is unclear which passages of the application as filed support the following features of operative Claim 2:

- (a) "(orienting means) for orienting at least a part the elongated particles substantially into the flow direction";
- (b) "with a passage opening which connects onto the outlets of the orienting means";
- (c) "wherein the form of the passage opening of the mould head (corresponds with the desired cross sectional form of the product)";
- (d) "(temperature curve) in the flow direction in the passage opening of the mould head".

- 
3. It is unclear which passages of the application as filed support the following features of operative Claim 4:
- (a) "(dimensioned relative to) the elongated particles";
  - (b) "... passage of the particles) longer than a certain predetermined length in a direction which corresponds other than to a dominating extent with the flow direction".
4. It is unclear which passage of the application as filed supports the following features of operative Claim 5:  
"shortest particles", longest particles".

To summarize, the operative set of claims is not considered to meet the requirements of Rule 70.2 (c) PCT.

4. Additional observations, if necessary:

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V. Reasoned statement under Article 35(2) with regard to novelty, inventive step and industrial applicability; citations and explanations supporting such statement

1. STATEMENT

Novelty (N)	Claims 1-7 _____	YES
	Claims _____	NO
Inventive Step (IS)	Claims 1-7 _____	YES
	Claims _____	NO
Industrial Applicability (IA)	Claims 1-7 _____	YES
	Claims _____	NO

2. CITATIONS AND EXPLANATIONS

The prior art cited in the International Search Report is silent on a method or an apparatus for manufacturing reinforced plastic-based composite products wherein a prismatic mould head having a specific temperature profile is employed.

Hence, claims pertaining to said method and apparatus which are in accordance with Rule 70.2(c) PCT are considered to meet the requirements of Art. 33 (2)-(4) PCT (cf. original claims 20-28).



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VII. Certain defects in the international application

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The following defects in the form or contents of the international application have been noted:

1. Although Claims 1 and 2 are drafted in the two-part form the feature "prismatic mould head" is incorrectly placed in the preamble, as it is not disclosed in US-A-5 082 605 (D2) or US-A-5 096 046 (Rule 6.3 (b) PCT).
2. In the first part of the example only ligneous fibrous particles having a length of 0.6 - 3 mm are employed. It is not clear how fibrous particles having a length of 1.9 mm adopt a random orientation when the fibrous particles having a length of 2 - 3 mm are "oriented to a dominant orientation" (i.e. > 50 %).

**INTERNATIONAL PRELIMINARY EXAMINATION REPORT**

Intern. application No.

PCT/NL95/00153

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**VIII. Certain observations on the international application**

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The following observations on the clarity of the claims, description, and drawings or on the question whether the claims are fully supported by the description, are made:

The meaning of features 1(b), 1(e) and 3(b) enumerated under point I.3. above is unclear (Art. 6 PCT).

values. Therefor, the application of the products which are made according to this known process are limited to ornamentation, covering, fence palings, etc.

- 5 The US patent US 5,096,406 describes an extruder assembly for composite materials, in particular materials comprising elongated wood particles which are embedded in a polymeric material. The composite material is extruded under high pressure, which can cause breaking of the particles. After  
10 the composite material leaves the mould head, the pressure on the material falls, which leads to the same drawback as the method according to the US patent 5,082,605.

- The object of the invention is a method and an apparatus to  
15 produce a composite product at low cost but which meets high standards for mechanical properties comparable to the properties of for example wood and also meets high standards for for instance fire resistance, weather resistance, non-aging properties and the like, wherein a large amount  
20 of particles can be applied without the risk of delaminations and the like.

- This object of the invention is achieved with a method wherein the particles have a length between 0.2 and about 6  
25 mm wherein the larger particles, which have a length of about 2 - 6 mm, are oriented to a dominant orientation while the smaller particles show a random orientation, and in that after entering the mould head the finished product has a temperature above the Vicat softening temperature of  
30 the applied plastic respectively the highest Vicat softening temperature of all the applied plastics, modelling the product to a desired shape, and cooling the product in this shape to below the said Vicat softening temperature while maintaining a chosen pressure. With this measure, a con-  
35 solidation of the composite material is achieved which prevents delamination and matrix failure. As a result, a composite material is obtained with mechanical properties which meet high standards.

## Method and apparatus for manufacturing a plastic based composite product

The invention relates to a method for manufacturing a plastic-based composite product which consists at least partially of a plastic in which a material consisting substantially of elongated particles is homogeneously embedded, wherein first the particles are mixed with the plastic and plasticized and subsequently at least a part of the particles is oriented, and wherein subsequently the mixture of the plastic and the particles passes a substantially prismatic mould head with a passage opening which connects onto the outlets of the orienting means, wherein the form of the passage opening of the mould head corresponds with the desired cross sectional form of the product. The invention also relates to an apparatus for carrying out such a method.

Such a method is known from the US patent US-A 5,082,605. This known method is particularly meant for producing a composite comprising wood particles which are embedded in a polymeric matrix. However, the properties of the composite are inferior in comparison with, for example, natural wood, since the polymeric matrix dominates the properties of the composite. The wood particles are relatively large and function as a filler. Therefor these wood particles hardly influence the mechanical properties, such as strength. The method as described in this US patent is an extrusion method. After leaving the extruder die, rollers are used to maintain dimensional stability of the extrudate. These rollers are adapted to exert only minimal pressure on the extrudate. This minimal pressure can only be exerted where the rollers are in contact with the extrudate. As a consequence the pressure on the composite material is very low and not uniform. With such extrusion methods an afterflow in the calibration unit is normally necessary to enable replenishing of the material deficit resulting from shrinkage. This after-flow causes delamination and a decrease in cohesion between the particles and the plastic. This results in mechanical properties with relatively low

The object of the invention is also achieved with an apparatus of the above mentioned type, wherein the length of the mould head and the temperature curve in the flow direction in the passage opening of the mould head are such  
5 that at the end of the passage opening the product has cooled to below its Vicat softening temperature. With such a apparatus it has become possible to cool the composite material in a controllable way under a certain pressure until below the Vicat softening temperature within a continuous process.  
10

With the method and the apparatus according to the invention, a composite material comprising particles which are embedded in a polymeric matrix, can be manufactured without  
15 waste of particles and, if desired, in a continuous process, which product does not only posses mechanical and physical/chemical properties corresponding with wood but can also be manufactured on the basis wood, for instance inferior wood, which can be classified as interior wood,  
20 exterior wood or construction wood.

The particles may be fibers or may be plates of wood or, for instance, mica.

25 Such composite material, which is manufactured according to the invention, will hereinafter be designated as technical wood. The obtained product can be applied at locations where tropical hardwood is still indicated.

30 Preferably, the plastic is a thermoplastic polymer material, in particular at least one polyolefin or one polymer on a basis of styrene, wherein  
a. the transverse dimension of the larger wood particles is preferably such that the ratio of the length in the  
35 fiber direction of the particles to this transverse dimension amounts to a minimum of 4, but preferably lies in the range of 6 - 80;  
b. The wood particles are present in a quantity of 40 - 80 % by mass, but preferably from 50 - 70 % by mass in

relation to the mass of product;

c. The obtained product complies minimally with the following requirements relating to mechanical properties in

- . bending strength in the fiber direction: 8 MPa
- 5 . bending modulus in the fiber direction: 3 GPa
- . tensile strength in the fiber direction: 6 MPa
- . tensile stress modulus in the fiber direction: 3 GPa
- . tensile strength transversely of fiber direction: 0.3 MPa
- 10 . tensile strength modulus transversely of fiber direction: 1 GPa

*continue page 2, line 35 of original text*

~~/material and the plastic is a thermoplastic polymer material~~  
in particular at least one polyolefin or one polymer on a basis of styrene, wherein

a. the transverse dimension of the large wood particles is preferably such that the ratio of the length in the principal direction of the particles to this transverse dimension amounts to a minimum of 4, but preferably lies in the range of 6-80.

b. the wood particles are present in a quantity of 40-80% by mass, but preferably from 50 to 70% by mass in relation to the mass of product.

c. the obtained product complies minimally with the following requirements relating to mechanical properties in

- . bending strength in the fibre direction: 8 MPa
- 15 . bending modulus in the fibre direction: 3 GPa
- . tensile strength in the fibre direction: 6 MPa
- . tensile stress modulus in fibre direction: 3 GPa
- . tensile strength transversely of fibre direction: 0.3 MPa
- . tensile stress modulus transversely of fibre direction: 1 GPa.

The invention herewith provides a product which in the manner to be described hereinbelow can be manufactured on a basis of wood without waste and, if desired, as a continuous process, which product not only possesses mechanical and physical/chemical properties corresponding with wood but which product can be manufactured on the basis of basic wood which can if desired be inferior and can be classified as interior wood, exterior wood or construction wood. This product according to the invention will hereinafter be designated as technical wood.

The invention provides the possibility of applying the obtained product at locations where tropical hardwood is still indicated.

For this purpose particles with tensile strength such as wood particles, preferably originating from waste wood or wood waste, optionally together with suitable strengthening particles, are predominantly oriented and bound by means of a thermoplastic polymer, preferably on a basis of olefins or styrene, wherein the proportion of applied particles is extreme

Bending tests (ISO 178):

Maximum bending load	32.0 MPa	0.3 MPa
Bending modulus	3.849 GPa	0.098 GPa

5

Addition of 10% glass or flax fibre with a length of 4 mm and L/D 150 to 400 gives values about 25% higher than shown above.

A product can in principle be manufactured in any  
10 suitable manner, starting from pre-compounded material or semi-manufacture, for instance by (isostatic) pressing, injection moulding, extruding, compushtruding.

A specific embodiment has the feature that the product consists substantially of a laminate comprising a  
15 plastic-based composite layer according to the above stated specifications; a first skin layer adhered thereto on one side and possessing chosen properties; optionally a second skin layer adhered thereto on the other side and possessing chosen properties; and which layers are  
20 mutually adhered by for instance glueing, welding, mirror-welding, with an infrared laser, with hot air or other suitable treatment.

Another embodiment is characterized by at least one additive for obtaining desired properties, which additive  
25 is added to the process flow at a suitable position in the compushtruding process during manufacture of the product.

The said chosen properties of the skin layers may relate for instance to the adherability of paint, lacquer  
30 and glue.

A variant of this latter embodiment has the feature that the skin layer or at least one of the skin layers is of the type according to the invention and the long particle orientation thereof has a chosen direction  
35 relative to this orientation of the first mentioned plastic layer. Specific mechanical properties can hereby be achieved. ~~/These can be based for instance on the stringer effect/~~



## CLAIMS

1. Method for manufacturing a plastic-based composite product which consists at least partially of a plastic in which a material consisting substantially of elongated particles is homogeneously embedded, wherein first the particles are mixed with the plastic and plasticized and subsequently at least a part of the particles is oriented to a flow direction, and wherein subsequently the mixture of the plastic and the particles passes a substantially prismatic mould head with a passage opening which connects onto the outlets of the orienting means, wherein the form of the passage opening of the mould head corresponds with the desired cross sectional form of the product **characterized in that**, the particles have a length between 0.2 and about 6 mm wherein the larger particles, which have a length of about 2 - 6 mm, are oriented to a dominant orientation while the smaller particles show a random orientation, and in that after entering the mould head the product has a temperature above the Vicat softening temperature of the applied plastic respectively the highest Vicat softening temperature of all the applied plastics, modelling the product to a desired shape, and cooling the product in this shape to below the said Vicat softening temperature while maintaining a chosen pressure.
2. Apparatus for manufacturing a plastic based composite product, which product consists at least partially of a plastic in which elongated particles are homogeneously embedded, which apparatus comprises:
  - means for plasticizing a mixture consisting substantially of the plastic and the particles, which particles are mixed with the plastic either beforehand or in the compounder, and pressing the plastic mixture to the outside via an outlet;
  - orienting means for orienting at least a part the elongated particles substantially into the flow direction;
  - a substantially prismatic mould head with a passage

opening which connects onto the outlets of the orienting means, wherein the form of the passage opening of the mould head corresponds with the desired cross sectional form of the product **characterized in that**, the length of the mould head and the temperature curve in the flow direction in the passage opening of the mould head are such that at the end of the passage opening the product has cooled to below its Vicat softening temperature.

10 3. Apparatus according to claim 2 **characterized in that** the plasticizing means consist of a compounder operating at low pressure and in that the apparatus further comprises a rotating displacement pump for further transporting the plastic mixture.

15 4. Apparatus as claimed in claim 2 or 3 **characterized in that**, the apparatus comprises a distribution head further guiding the mixture in order to further transport the plastic mixture substantially as a plug flow, wherein the orienting means comprise at least one bundle of substantially parallel channels through which the plastic mixture can flow, which channels are dimensioned relative to the elongated particles such that they are too small to allow passage of the particles longer than a certain predetermined length in a direction which corresponds other than to a dominating extent with the flow direction.

5. Apparatus as claimed in one of the preceding claims 2 - 4 **characterized in that**, the linear transverse dimensions of at least the parts of the channels located furthest downstream amount to a minimum of 2x the length of the shortest particles and a maximum of 1.5x the length of the longest particles.

35 6. Apparatus as claimed in one of the preceding claims 2 - 5 **characterized in that**, the orienting means comprises at least two bundles of channels mutually connecting in series, wherein the transverse dimensions of the channels of a bundle located further downstream are smaller than

those of the channels of a bundle located further upstream.

7. Apparatus as claimed in one of the preceding claims 2  
5 - 6 characterized in that, at least one feed for adding  
particle material and/or additives to the plastic connects  
to the compounder.

1A 405 Rec'd PCT/PTO 27 OCT 1998

values. Therefor, the application of the products which are made according to this known process are limited to ornamentation, covering, fence palings, etc.

- 5 The US patent US 5,096,406 describes an extruder assembly for composite materials, in particular materials comprising elongated wood particles which are embedded in a polymeric material. The composite material is extruded under high pressure, which can cause breaking of the particles. After  
10 the composite material leaves the mould head, the pressure on the material falls, which leads to the same drawback as the method according to the US patent 5,082,605.

- The object of the invention is a method and an apparatus to  
15 produce a composite product at low cost but which meets high standards for mechanical properties comparable to the properties of for example wood and also meets high standards for for instance fire resistance, weather resistance, non-aging properties and the like, wherein a large amount  
20 of particles can be applied without the risk of delaminations and the like.

- This object of the invention is achieved with a method wherein the particles have a length between 0.2 and about 6  
25 mm wherein the larger particles, which have a length of about 2 - 6 mm, are oriented to a dominant orientation while the smaller particles show a random orientation, and in that after entering the mould head the finished product has a temperature above the Vicat softening temperature of  
30 the applied plastic respectively the highest Vicat softening temperature of all the applied plastics, modelling the product to a desired shape, and cooling the product in this shape to below the said Vicat softening temperature while maintaining a chosen pressure. With this measure, a con-  
35 solidation of the composite material is achieved which prevents delamination and matrix failure. As a result, a composite material is obtained with mechanical properties which meet high standards.

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## CLAIMS

1. Method for manufacturing a plastic-based composite product which consists at least partially of a plastic in which a material consisting substantially of elongated particles is homogeneously embedded, wherein first the  
5 particles are mixed with the plastic and plasticized and subsequently at least a part of the particles is oriented to a flow direction, and wherein subsequently the mixture of the plastic and the particles passes a substantially prismatic mould head with a passage opening which connects  
10 onto the outlets of the orienting means, wherein the form of the passage opening of the mould head corresponds with the desired cross sectional form of the product **characterized in that**, the particles have a length between 0.2 and about 6 mm wherein the larger particles, which have a  
15 length of about 2 - 6 mm, are oriented to a dominant orientation while the smaller particles show a random orientation, and in that after entering the mould head the product has a temperature above the Vicat softening temperature of the applied plastic respectively the highest Vicat softening temperature of all the applied plastics, modelling the  
20 product to a desired shape, and cooling the product in this shape to below the said Vicat softening temperature while maintaining a chosen pressure.

25 2. Apparatus for manufacturing a plastic based composite product, which product consists at least partially of a plastic in which elongated particles are homogeneously embedded, which apparatus comprises:

- means for plasticizing a mixture consisting substantially of the plastic and the particles, which particles  
30 are mixed with the plastic either beforehand or in the compounder, and pressing the plastic mixture to the outside via an outlet;
- orienting means for orienting at least a part the  
35 elongated particles substantially into the flow direction;
- a substantially prismatic mould head with a passage

opening which connects onto the outlets of the orienting means, wherein the form of the passage opening of the mould head corresponds with the desired cross sectional form of the product **characterized in that**, the length of the mould head and the temperature curve in the flow direction in the passage opening of the mould head are such that at the end of the passage opening the product has cooled to below its Vicat softening temperature.

10 3. Apparatus according to claim 2 **characterized in that** the plasticizing means consist of a compounder operating at low pressure and in that the apparatus further comprises a rotating displacement pump for further transporting the plastic mixture.

15 4. Apparatus as claimed in claim 2 or 3 **characterized in that**, the apparatus comprises a distribution head further guiding the mixture in order to further transport the plastic mixture substantially as a plug flow, wherein the orienting means comprise at least one bundle of substantially parallel channels through which the plastic mixture can flow, which channels are dimensioned relative to the elongated particles such that they are to small to allow passage of the particles longer than a certain predetermined length in a direction which corresponds other than to a dominating extent with the flow direction.

25 5. Apparatus as claimed in one of the preceding claims 2 - 4 **characterized in that**, the linear transverse dimensions of at least the parts of the channels located furthest downstream amount to a minimum of  $2x$  the length of the shortest particles and a maximum of  $1.5x$  the length of the longest particles.

35 6. Apparatus as claimed in one of the preceding claims 2 - 5 **characterized in that**, the orienting means comprises at least two bundles of channels mutually connecting in series, wherein the transverse dimensions of the channels of a bundle located further downstream are smaller than

those of the channels of a bundle located further upstream.

7. Apparatus as claimed in one of the preceding claims 2  
5 - 6 characterized in that, at least one feed for adding  
particle material and/or additives to the plastic connects  
to the compounder.

## CLAIMS

1. Method for manufacturing a plastic-based composite product which consists at least partially of a plastic in which a material consisting substantially of elongated particles is homogeneously embedded, wherein first the particles are mixed with the plastic and plasticized and subsequently at least a part of the particles is oriented to a flow direction, and wherein subsequently the mixture of the plastic and the particles passes a substantially prismatic mould head with a passage opening which connects onto the outlets of the orienting means, wherein the form of the passage opening of the mould head corresponds with the desired cross sectional form of the product **characterized in that**, the particles have a length between 0.2 and about 6 mm wherein the larger particles, which have a length of about 2 - 6 mm, are oriented to a dominant orientation while the smaller particles show a random orientation, and in that after entering the mould head the finished product is heated to a temperature above the Vicat softening temperature of the applied plastic respectively the highest Vicat softening temperature of all the applied plastics, modelling the product to a desired shape, and cooling the product in this shape to below the said Vicat softening temperature while maintaining a chosen pressure.
2. Apparatus for manufacturing a plastic based composite product, which product consists at least partially of a plastic in which elongated particles are homogeneously embedded, which apparatus comprises:
- means for plasticizing a mixture consisting substantially of the plastic and the particles, which particles are mixed with the plastic either beforehand or in the compounder, and pressing the plastic mixture to the outside via an outlet;
  - orienting means for orienting at least a part the elongated particles substantially into the flow direction;
  - a substantially prismatic mould head with a passage



opening which connects onto the outlets of the orienting means, wherein the form of the passage opening of the mould head corresponds with the desired cross sectional form of the product **characterized in that**, the length of the mould head and the temperature curve in the flow direction in the passage opening of the mould head are such that at the end of the passage opening the product has cooled to below its Vicat softening temperature.

10 3. Apparatus according to claim 2 **characterized in that** the plasticizing means consist of a compounder operating at low pressure and in that the apparatus further comprises a rotating displacement pump for further transporting the plastic mixture.

15

4. Apparatus as claimed in claim 2 or 3 **characterized in that**, the apparatus comprises a distribution head further guiding the mixture in order to further transport the plastic mixture substantially as a plug flow, wherein the orienting means comprise at least one bundle of substantially parallel channels through which the plastic mixture can flow, which channels are dimensioned relative to the elongated particles such that they are too small to allow passage of the particles longer than a certain predetermined length in a direction which corresponds other than to a dominating extent with the flow direction.

5. Apparatus as claimed in one of the preceding claims 2 - 4 **characterized in that**, the linear transverse dimensions of at least the parts of the channels located furthest downstream amount to a minimum of 2x the length of the shortest particles and a maximum of 1.5x the length of the longest particles.

35 6. Apparatus as claimed in one of the preceding claims 2 - 5 **characterized in that**, the orienting means comprises at least two bundles of channels mutually connecting in series, wherein the transverse dimensions of the channels of a bundle located further downstream are smaller than

those of the channels of a bundle located further upstream.

7. Apparatus as claimed in one of the preceding claims 2  
5 - 6 characterized in that, at least one feed for adding  
particle material and/or additives to the plastic connects  
to the compounder.

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<b>(21) International Application Number:</b> PCT/NL95/00153 <b>(22) International Filing Date:</b> 27 April 1995 (27.04.95) <b>(71) Applicant (for all designated States except US):</b> AVIPLAST B.V. [NL/NL]; Jutestraat 12, NL-7461 TR Rijssen (NL). <b>(72) Inventors; and</b> <b>(75) Inventors/Applicants (for US only):</b> VAN DIJK, Dirk [NL/NL]; Raalterweg 30, NL-7451 KW Holten (NL). DE VRIES, Fransiscus, Bernardus, Antonius [NL/NL]; Scholtenhofmansbrink 1, NL-7152 JJ Eibergen (NL). <b>(74) Common Representative:</b> SCHUMANN, Bernard, Herman, Johan; Arnold & Siedsma, Sweelinckplein 1, NL-2517 GK The Hague (NL).		<b>(81) Designated States:</b> AM, AT, AU, BB, BG, BR, BY, CA, CH, CN, CZ, DE, DK, EE, ES, FI, GB, GE, HU, IS, JP, KE, KG, KP, KR, KZ, LK, LR, LT, LU, LV, MD, MG, MN, MW, MX, NO, NZ, PL, PT, RO, RU, SD, SE, SG, SI, SK, TJ, TM, TT, UA, UG, US, UZ, VN, ARIPO patent (KE, MW, SD, SZ, UG), European patent (AT, BE, CH, DE, DK, ES, FR, GB, GR, IE, IT, LU, MC, NL, PT, SE), OAPI patent (BF, BJ, CF, CG, CI, CM, GA, GN, ML, MR, NE, SN, TD, TG).  <b>Published</b> <i>With international search report. In English translation (filed in Dutch).</i>
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<b>(57) Abstract</b> <p>The invention relates to a plastic-based composite product which consists at least partially of a plastic in which a material consisting substantially of particles is homogeneously embedded, which particles have tensile strength in at least one principal direction. The invention provides a product of the stated type which has the special feature that the particles comprise: small particles, in particular plates or fibres, with a random orientation and a length of 0.2-2 mm; and large particles with a dominant orientation, for instance 80-95 %, of the said principal direction of the particles in a chosen product principal direction and a length in the particle principal direction of about 2-6 mm.</p> <p style="font-size: 2em; transform: rotate(-10deg); opacity: 0.5;">Priority Document</p>		

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**PLASTIC-BASED COMPOSITE PRODUCT AND METHOD  
AND APPARATUS FOR MANUFACTURING SAME**

The invention relates to a plastic-based composite product which consists at least partially of a plastic in which a material consisting substantially of particles is homogeneously embedded, which particles have tensile strength  
5 in at least one principal direction.

It is an object of the invention to provide a product which can be manufactured at low cost but which nevertheless meets high standards which may be required in respect of diverse properties. Such properties can relate for instance  
10 to mechanical properties, fire resistance, weather resistance, non-ageing properties and the like.

With a view to the above, the invention provides a product of the stated type which has the special feature that the particles comprise:

15 small particles, in particular plates or fibres with a random orientation and a length of 0.2-2 mm; and

large particles with a dominant orientation, for instance 80-95%, of the said principal direction of the particles in a chosen product principal direction and a  
20 length in the particle principal direction of about 2-6 mm.

A specific embodiment has the characteristic that the large particles comprise plates and the particle principal direction extends in the main plane thereof.

In order to easily obtain the desired orientation of  
25 the plates the embodiment is recommended wherein the plates have an at least more or less isotropic tensile strength in their main plane.

Very suitable in this latter context is the variant wherein the plates consist substantially of mica.

30 It is noted that according to the invention the particles can also possess a generally elongate form, whereby they may be designated as fibres.

A specific and very advantageous variant has the feature that the particles consist predominantly of wood

material and the plastic is a thermoplastic polymer material, in particular at least one polyolefin or one polymer on a basis of styrene, wherein

- a. the transverse dimension of the large wood particles is preferably such that the ratio of the length in the principal direction of the particles to this transverse dimension amounts to a minimum of 4, but preferably lies in the range of 6-80.
- b. the wood particles are present in a quantity of 40-80% by mass, but preferably from 50 to 70% by mass in relation to the mass of product.
- c. the obtained product complies minimally with the following requirements relating to mechanical properties in
  - . bending strength in the fibre direction: 8 MPa
  - 15 . bending modulus in the fibre direction: 3 GPa
  - . tensile strength in the fibre direction: 6 MPa
  - . tensile stress modulus in fibre direction: 3 GPa
  - . tensile strength transversely of fibre direction: 0.3 MPa
  - . tensile stress modulus transversely of fibre direction:  
20 1 GPa.

The invention herewith provides a product which in the manner to be described hereinbelow can be manufactured on a basis of wood without waste and, if desired, as a continuous  
25 process, which product not only possesses mechanical and physical/chemical properties corresponding with wood but which product can be manufactured on the basis of basic wood which can if desired be inferior and can be classified as interior wood, exterior wood or construction wood. This product  
30 according to the invention will hereinafter be designated as technical wood.

The invention provides the possibility of applying the obtained product at locations where tropical hardwood is still indicated.

35 For this purpose particles with tensile strength such as wood particles, preferably originating from waste wood or wood waste, optionally together with suitable strengthening particles, are predominantly oriented and bound by means of a thermoplastic polymer, preferably on a basis of olefins or  
40 styrene, wherein the proportion of applied particles is extreme

relative to the binder polymer such that there is no question of this being a fibre-filled thermoplastic material but an exceptionally wood-like material which can be processed in a manner usual for an expert.

5           It is important that the method can be performed continuously and comprises the stages of compressing, compounding, fibre-orienting and shaping, wherein the following requirements must be fulfilled.

- 10           1.     the compression stage wherein expulsion of included gases, both inter- and intrafibular, must likewise take place under conditions wherein the fibres remain intact.
2.     the compounding phase wherein the mixing of components and wetting with polymer melt must take place under conditions wherein fibres sensitive to breakage and  
15           splitting remain intact.
3.     the orienting phase wherein a dominant orientation of long particles in the compressed plastic material must take place under conditions such that the mutual position of the particles in the mix is by and large retained,  
20           that is, the orientation proceeds along fluent flow lines, gradually and without such high shear stresses that there would thereby be a danger of thermal degradation.
4.     the shaping phase wherein the mix acquires its fixed  
25           shape and dimensioning on the cross section perpendicularly of the flow direction, likewise under conditions wherein the relative mutual position of the particles remains practically unchanged, for which purpose the material flows under overpressure out of the  
30           mould head and remains at pressure in the calibrating unit until the binding polymer phase has cooled to below the Vicat softening temperature.

          There is no extrusion here, since in extrusion the melted mass, after leaving the extrusion head, is relatively  
35   pressureless or is at least under a relatively small overpressure such as is necessary to allow an after-flow of the plastic core of the shaped product in the calibrating unit in order to thus enable replenishing of the material deficit resulting from shrinkage. This after-flow must always be



avoided because it would be accompanied by delamination and a decrease in cohesion between particles and plastic and the desired properties would consequently not be obtained.

There is also no question of pulltrusion since tensile  
5 forces are therein exerted on the shaped product after and in the final part of the consolidation phase for the purpose of continuous discharge of the shaped product, which otherwise acquires its shape in pressureless manner or under relatively small overpressure from a fusing of thermoplastic and fibre  
10 reinforcement.

The method according to the invention involves a shaping process under continuous significant overpressure, wherein the material does not flow but is pushed forward as a plastic plug through orientation, shaping and consolidation phase.

15 This method is therefore designated with the name of pushtrusion.

The obtained product on the basis of wood particles and plastic, technical wood, is fully recyclable due to the fibre binding by means of a thermoplastic. It will be apparent that  
20 recycled polyolefins and styrenes can also be used as fibre binder.

Additives can moreover be added during the compounding phase, whereby properties are given to the technical wood which surpass those of natural wood, particularly when fast-growers  
25 such as firs, pines, birches and poplars are used as starting point.

The method as continuous process will be designated as compushtrusion and the apparatus in which the method is performed will be designated compushtruder.

30 Examples are known from the patent literature wherein plastic products resembling wood are manufactured but wherein the mechanical properties as obtained according to the method of the invention are not obtained therein.

The known methods, insofar as a relatively high  
35 proportion of wood mass is processed into the product, moreover make use without exception of a machine with a strong kneading action, such as for instance the Banbury mixer or the Buss

Co-kneader, which is deliberately not used in the new method in respect of the harmful effect of such a kneading machine on the dimensions and properties of the applied wood fibres.

The applications of the products according to the known  
5 methods are limited to ornamentation, covering, fence palings etc., wherein the wood part is considered as filler:

EP-A-0 114 409, E.I. Du Pont de Nemours, 1994

US-A-5 030 662, A.K. Banerjee, 1991

US-A-4 866 110, Chang Y. Lee, 1989

10 US-A-5 082 605, J.G. Brooks et al, 1992

NL-A-77 04265, Lankhorst Touwfabrieken, 1978

WO-A-90/08020, Polywood Patent AB, 1990.

Description of the construction of the compushtruder.

15

On the compushtruder can be distinguished:

- (1) compounder: preferably twin-screw or single-screw with adapted screw geometry,
- 20 (2) rotating displacement pump, preferably toothed wheel, plunger or screw,
- (3) distribution head
- (4) orienting means,
- (5) mould head, also calibrating device,

25

which parts are further described hereinafter.

(1) Compounder.

The function of the compounder, as for instance from the  
30 series Theysohn TSK-N, Leistritz ZSE-GL, is to encapsulate particles, in particular wood fibres, in the liquid thermoplastic. This must take place such that the wood fibre is completely enclosed with polymer, however without the fibres effectively being shortened herein through breaking in the  
35 length. The wood fibres may however split longitudinally. This situation is achieved by the polymer melt wetting the fibres. Due to the relatively strong adhesion properties of the melt in relation to the fibres the latter are carried along and dispersed homogeneously further on in the melt phase. Adapting

the screw geometry prevents too intensive a kneading action being applied to the mixture, as is usually the case with for instance the Banbury mixer or a standard compounder. A conscious choice has been made for a long compounding path  
5 instead of a short sharp one, wherein the wood particles are handled carefully while a homogeneous dispersion nevertheless occurs. The wood particles are preferably introduced under compression and compacted to prevent excess air inclusion which could affect the quality of the end product.

10 By successively compressing, decompressing and degassing under vacuum the mixtures by means of the chosen screw geometry, air and gases are expelled, both from the mixture and from the pores in the fibres. In the case of a single-screw compounder the mixture is driven in a compressed plug flow,  
15 whereafter the thermoplastic part is melted.

The adhesion between wood particles and polymer melt is so great and adheres such that optional further additions can be made to the mixture without breaking the adhesion between polymer and wood particles.

20 Optional reinforcing fibres on a basis of cellulose, such as flax, hemp or mineral fibres of glass, (mica) plates or the like can be carried along in the above path.

The pressure on the mixture for the purpose of the shaping phase is not provided, as is usual, by the principle of  
25 the extruding action of the screw but by a rotating displacement pump interposed for this purpose.

## (2) Rotating displacement pump

The pressure required to press the mixture through the  
30 distribution head (to be described below), the orienting means and the mould head/calibrating device is produced by a displacement pump, preferably a screw pump or a gear pump, for example Maag, Expac type Estrex 56/56 or Witte type ESTHF 92.6 LKK. High pressures can be realized without any significant  
35 effect on particle shape and dimensions. Moreover, since large shearing forces are practically absent, the desired pressure increase can be realized without excessive thermal load and degradation of the mixture.

### (3) Distribution head

The distribution head is a necessary "interface" between rotating displacement pump and orienting means. The laminar flow pattern from the rotating displacement pump must be converted to a plug flow.

The particle orienting means must be supplied with a constant flow of mixture which in quantity and speed is distributed equally over the flow surface of the orienting means, this in order to prevent blockage and turbulence as result of speed differences after each phase of the particle orientation. The distribution head thus initiates the required placidity within the flow profile. The plug flow can be optimized:

- \* mechanically by varying the through-flow surface per channel using adjustment bolts which can be accessed from outside and/or
- \* thermally by varying the flow resistance per channel using heating elements.

The form of the distribution head depends partly or wholly on the desired product.

### (4) Orienting means

The orienting means consist of a plurality of plates connected in series with slits (in the case of plates) or (cylindrical) holes (in the case of fibres) in the flow direction. The total through-flow surface per plate is mutually equal. The diameter of the holes decreases over the plates connected in series. The diameter and the decrease thereof over the plates depends on the particle dimensions and the desired degree of orientation. When short fibres with a length of 2 mm and an L/D ratio of 4 are guided through a hole of 2.5 mm, little or no orientation will occur.

If a long fibre with a length of 4 mm and an L/D ratio of 8 passes through the same hole, then orientation will occur.

The number of plates depends on the degree of uniformity in the fibre length, the L/D ratios thereof and the desired degree of orientation.

The length of the slits or cylindrical holes is preferably at least equal to the maximum linear particle dimension.

Transition plates between the hole plates with different diameters have of course the same through-flow surface as the hole plates.

(5) Mould head/calibrating device

In order to make a two-dimensionally shaped endless product from the "wood mixture" with the oriented particles a mould head and a calibrating device are necessary. In contrast to the typically used extrusion shaping process, mould head and calibrating device are constructed integrally in the compushtrusion process, since the pushtrudate may not be pulled or have pressure exerted thereon in lengthwise direction.

An essential condition for obtaining technical wood is that after shaping the mixture is cooled under pressure to below the Vicat softening temperature of the binder thermoplastic. This consolidation process is necessary to prevent delamination and matrix failure.

The system pressure is derived from the flow resistance between mixture and mould head/calibration.

In order to prevent delamination and matrix failure in the calibration the first part hereof is covered with a coating which substantially reduces the surface resistance to a value wherein no delamination and matrix failure occurs. The outer part of the product meanwhile cooled to below the Vicat softening temperature in the calibration provides matrix support of that part of the product, the material core, still lying below the Vicat softening temperature.

Process parameters

It will be apparent that in the compushtrusion process many parameters can be distinguished which all have their influence on the creation, quality and properties of the technical wood.

The process parameters can be sub-divided as follows:

- (1) Variable parameters, being adjustable parameters which can be varied on-line.
- (2) Adaptable parameters are set once subject to the type of technical wood and the desired product section.

5

(1) Variable parameters

The variable parameters can be further sub-divided into two categories:

10

(1)a. as they also occur in the known compounding/extrusion process:

- temperatures, for instance set temperature profile of the screw cylinder, temperature of mould head,
- 15       - screw rotation speed,
- with twin-screw compounder: the filling level by means of dosaging,
- pressure, interactive,
- degassing, absolute pressure of the vacuum.

20

(1)b. new compushtrusion parameters:

- through-flow surface adjusted by means of adjustment bolts and/or temperature profile of the channels in the distribution head,
- 25       - temperature of the particle orienting provision,
- temperature of the calibration,
- rotating displacement pump temperature,
- rotating displacement pump pressure and output.

30

(2) Adaptable parameters

- screw geometry,
- manner and position of dosage,
- degassing zones and number,
- 35       - L/D ratio of the compushtruder,
- number of orientation plates,
- diameters and lengthwise change in the holes in the orientation plates,
- flow resistance in the mould head/calibrating device,

- position and active length of the resistance-reducing coating in the calibrating device.

Example of the method and the mechanical properties of obtained  
5 technical wood.

The method is performed in a compushtruder as described above  
with a capacity of 200 kg per hour.

10 The composition of the mixture is as follows:

- 60% by mass pinewood, length of the fibrous particles  
0.6 - 3 mm,  
L/D = 4, moisture content 2.2% by mass
- 40% by mass polypropylene. MFI = 15 dg/min, (230/2,16)
- 15 - no other additions.

The most important process parameters are as follows:

- temperature profile in the range 160-195°C
- 20 - compounder feed pressure 15 bar
- rotating displacement pump pressure 95 bar
- vacuum 20 kPa.

Measured mechanical properties of the technical wood:

25

Six samples were measured wherein the following results  
were obtained in respect of the length orientation of the  
wood particles:

30 Tensile strength tests (ISO 527) standard deviation

	Maximum tensile force	23.5 MPa	0.4 MPa
	E-modulus	5.737 GPa	0.104 GPa
35 Tensile strength			
	transversely of fibre direction	12.2 MPa	0.35 MPa
	Tensile stress modulus		
	transversely of fibre direction	2.122 GPa	0.102 GPa

Bending tests (ISO 178):

Maximum bending load	32.0 MPa	0.3 MPa
Bending modulus	3.849 GPa	0.098 GPa

5

Addition of 10% glass or flax fibre with a length of 4 mm and L/D 150 to 400 gives values about 25% higher than shown above.

10 A product can in principle be manufactured in any suitable manner, starting from pre-compounded material or semi-manufacture, for instance by (isostatic) pressing, injection moulding, extruding, compushtruding.

15 A specific embodiment has the feature that the product consists substantially of a laminate comprising a plastic-based composite layer according to the above stated specifications; a first skin layer adhered thereto on one side and possessing chosen properties; optionally a second skin layer adhered thereto on the other side and possessing chosen properties; and which layers are  
20 mutually adhered by for instance glueing, welding, mirror-welding, with an infrared laser, with hot air or other suitable treatment.

Another embodiment is characterized by at least one additive for obtaining desired properties, which additive  
25 is added to the process flow at a suitable position in the compushtruding process during manufacture of the product.

The said chosen properties of the skin layers may relate for instance to the adherability of paint, lacquer  
30 and glue.

A variant of this latter embodiment has the feature that the skin layer or at least one of the skin layers is of the type according to the invention and the long particle orientation thereof has a chosen direction  
35 relative to this orientation of the first mentioned plastic layer. Specific mechanical properties can hereby be achieved. These can be based for instance on the Stringer effect.



As already noted, the product according to the invention lends itself very well for manufacture by means of a process which can best be described as compushtrusion. In this respect the invention likewise  
5 provides a compushtrusion apparatus for manufacturing a product of the above specified type, which apparatus comprises: a compounder operating at low pressure for plasticizing a mixture consisting substantially of plastic and particles with tensile strength, which  
10 particles are mixed with the plastic either beforehand or in the compounder, and pressing the plastic mixture to the outside via an outlet;

a rotating displacement pump further transporting the plastic mixture;

15 a distribution head further guiding the mixture in order to further transport the plastic mixture substantially as a plug flow;

orienting means further guiding the mixture, comprising at least one bundle of substantially parallel  
20 channels through which the plastic mixture can flow and which are dimensioned relative to the long particles such that, other than to a dominating extent in the particle principal direction, they are too small to allow passage of the long particles present in the plastic mixture; and

25 a substantially prismatic mould head which connects onto the outlets of the channels and the form of which corresponds with the desired cross sectional form of the product;

such that the principal direction of the particles  
30 corresponds with the longitudinal direction of the mould head and the principal direction of the product;

which mould head is so long and has a temperature curve in the longitudinal direction such that at the end of the mouth the product has cooled to below its Vicat  
35 softening temperature.

As already discussed, the adhesion of lacquer, paint and glue can be enhanced by mechanical means, for instance due to a certain porosity and protruding fibres. Another embodiment which achieves the same effect in

chemo-physical manner is characterized by an additive with a desired influence on chosen properties of the product and belonging to at least one of the following classes:

- 5           . influencers of adhesion between particles with tensile strength and matrix polymer (class H),
- . influencers of the properties of the surface of the product, particularly in respect of coatings or adhesives for applying in sandwich structures
- 10           (class O),
- . influencers of the pyrogenic properties (class P),
- . influencers of the particle durability (class D),
- 15           . blowing means for obtaining a foamed structure (class B), in the case of a sufficiently high temperature increase.

The invention will now be elucidated with reference to the annexed drawings, wherein:

- 20           Figure 1 shows a highly schematic, partly broken away perspective side view of a compushtrusion apparatus with which a product according to the invention can be manufactured;

- Figure 2 shows a highly schematic side view of an
- 25           apparatus with which another product according to the invention can be manufactured;

Figure 3 is a cut away perspective view corresponding with figure 1 of a variant; and

- Figure 4 is a cut away perspective view on enlarged
- 30           scale of a part of the apparatus according to figure 3.

- Figure 1 shows a compushtruder 1. This comprises a compounder 2, a rotating displacement pump 3, a distribution head 4, a particle orienting member 42 and a mould head/calibrating unit 43. The distribution head 4
- 35           and the orienting head 5 comprise continuous channels through which can pass the plasticized plastic, in which fibres with tensile strength are incorporated. The long fibres have a length predominantly in the range of 2-6 mm. In order to give these fibres the desired

orientation, that is, the longitudinal direction designated with arrow 6, the continuous channels comprise in the distribution head 4 and the orienting head 5 sub-channels having an effective diameter of for instance 5 in the order of 6-8 mm, taking account of the wish to choose the diameter size of the said channels such that the orientation of the short fibres is not affected. It will be apparent that for other fibre lengths the said passages can be adapted correspondingly. The product 7 consists substantially of the solidified composite mass 10 in which the long fibres extend generally at least roughly in longitudinal direction 6.

Figure 2 shows an alternative. Two compushtruders 8, 9 deliver products 10, 11 respectively in the 15 direction of the pinch between two rollers 12, 13. At the position of the pinch 14 a plastic-impregnated fibre mat 15, comprising for instance glass or aramide fibres, is introduced between the plate-like products 10, 11 by transport rollers 16, 17. A laminate 18 is thus formed by 20 heating and by the force exerted by rollers 12, 13. The fibres in the fibre mat 15 extend substantially in the longitudinal direction 6. At least one of the products 10, 11 displays a desired porosity in order to ensure properties corresponding with wood.

25 For greater thicknesses a product can be manufactured from a plurality of laminates placed one on top of another and mutually adhered by a thermal treatment and pressure.

It is further noted in general that binders can 30 also be added to the basic plastic in order to improve the adhesion between fibres, plastic and other additives. The adhesion to the product according to the invention of paints and lacquers on acryl-water basis can hereby also improve.

35 The use of colouring agents or pigments in the mass can provide the advantage that a uniform product is obtained.

With reference to figure 2 attention is drawn to the fact that by making use of for example co-extrusion

or other suitable technique on the product according to the invention an additional coating can be applied to the visual side of the product. Such a coating can have an extra-stable colour, an increased UV-resistance or  
5 resistance to dirt and weather influences.

In contrast to the above mentioned prior art the invention provides a product which lends itself for processing and treating as wood. The following considerations are paramount here.

10 The product according to the invention can have a linear expansion similar to wood and also has a similar strength and stiffness with a comparatively great toughness and excellent cracking strength. The product can display fire characteristics which, by making use of  
15 environmentally-friendly provisions, must be at least the equal of the fire characteristics of normal wood and which when it burns may not produce any more smoke and harmful substances than normal wood. At a specific weight of  $750-1250 \text{ kg/m}^3$ , the process and the required raw  
20 materials and additives may not exceed the price of normally processed wood.

In terms of appearance and weight the product according to the invention can if desired display a striking similarity with natural wood or traditionally  
25 pressed wood such as MDF or wood fibre board.

The product according to the invention can be worked with normal tools and normal wood processing machines. In the usual manner of wood it can be nailed, sawn, screwed, glued, painted and lacquered.

30 The product according to the invention can possess an excellent resistance to climatological conditions such as moisture, sunlight with ultraviolet component, temperature changes etc.

The products according to the invention are better  
35 suitable for recycling after use than natural wood.

The products according to the invention are superior to wood in respect of moisture absorption, rotting, swelling and the like.

Figure 3 shows a compushtruder 21 which has a structure differing partially from that drawn in figure 1. A distribution head 23 connects onto the rotating displacement pump 3 (see also figure 4). This block has a more or less conically tapering inlet space 24 which debouches into a fan of nine channels 25-33. Figure 4 in particular shows clearly the internal structure of the distribution head 23. By means of screws controllable from outside, which are all designated 34 for the sake of convenience, the effective passage and therewith the flow resistance of channels 25-33 can be individually adjusted. An excellent homogeneity of the through-flowing mixture can hereby be ensured. Alternatively, the effective through-flow of the channels can be influenced by a selective heating. The through-flow of a channel can in any case be improved by reducing the flow resistance. This can be achieved not only by adjusting the passage but also by changing the temperature, whereby the viscosity of the through-flowing mixture changes, at least in the boundary surface, thereby changing the effective through-flow.

Via a collection space 35 the plastic mixture allowed through is fed to a bundle 36 of channels which is bounded by plates extending one on top of another in transverse direction. By means of channels 37 sufficient heat can be supplied to the through-flowing mixture to keep it in plastic state. A second bundle 38 follows the first bundle 36 via an interspace 37. A third bundle 40 follows via an interspace 39.

Downstream of this latter bundle 40 is situated the mould head 41 of the compushtruder 21. By means of calibrating and cooling means the product is then cooled gradually while a sufficient pressure is maintained, and is carried to the outside in the correct dimensions. Product 42 according to figure 3 has an orientation of the long fibres corresponding with the mould longitudinal direction.

The diameters of the mutually adjacent round

channels in bundles 36, 38, 40 amount in this embodiment to respectively 14 mm, 8 mm and 6 mm while retaining the same through-flow surface.

5 It will be apparent from the structure shown in figure 4 that the mass flowing through the more inwardly located channels has a smaller flow resistance than the mass flowing through the more outwardly located channels, since they are longer. In this context the more inwardly located channels can have a slightly smaller diameter.

10 It is noted that in all compushtruders the mixture with the fibres embedded therein is introduced into the mould head by the rotating displacement pump at a relatively high temperature above the Vicat softening temperature, for instance 180°C, cools during the  
15 transport through the mould head and has reached a reduced temperature at the end such that the exiting product has a temperature below the Vicat softening temperature, whereby it is sufficiently cured that it will no longer undergo any substantial change in shape.

\*\*\*\*\*

**CLAIMS**

1. Plastic-based composite product which consists at least partially of a plastic in which a material consisting substantially of particles is homogeneously embedded, which particles have tensile strength in at least one principal direction,  
5 characterized in that  
the particle comprise:  
small particles, in particular plates or fibres with a random orientation and a length of 0.2-2 mm; and  
10 large particles with a dominant orientation, for instance 80-95%, of the said principal direction of the particles in a chosen product principal direction and a length in the particle principal direction of about 2-6 mm.
- 15 2. Product as claimed in claim 1,  
characterized in that  
the large particles are plates and the particle principal direction extends in the main plane thereof.
- 20 3. Product as claimed in claim 2,  
characterized in that  
the plates have an at least more or less isotropic tensile strength in their main plane.
- 25 4. Product as claimed in claim 3,  
characterized in that  
the plates consist substantially of mica.
5. Product as claimed in claim 1,  
characterized in that  
the large particles are fibres wherein the principal direction of the particles is the longitudinal  
30 direction of each fibre.
6. Product as claimed in claim 1,  
characterized in that  
the particles consist predominantly of wood material and the plastic is a thermoplastic polymer

material, in particular at least one polyolefin or one polymer on a basis of styrene,

wherein

- a. the transverse dimension of the large wood particles is preferably such that the ratio between the length in the principal direction of the particles and this transverse dimension amounts to a minimum of 4, but preferably lies in the range of 6-80.
- b. the wood particles are present in a quantity of 40-80% by mass, but preferably from 50 to 70% by mass in relation to the mass of product.
- c. the obtained product complies minimally with the following requirements relating to mechanical properties in
  - 15 . bending strength in the fibre direction: 8 MPa
  - . bending modulus in the fibre direction: 3 GPa
  - . tensile strength in the fibre direction: 6 MPa
  - . tensile stress modulus in fibre direction: 3 GPa
  - . tensile strength transversely of fibre direction: 0.3 MPa
  - 20 . tensile stress modulus transversely of fibre direction: 1 GPa.
7. Product as claimed in claim 6, characterized in that the wood particles originate from softwood or hardwood, preferably one of the following types: fir, spruce, birch, poplar.
8. Product as claimed in claim 6, characterized by other particles with tensile strength of 3-25% by mass, preferably 5-18% by mass.
9. Product as claimed in claim 8, characterized by other particles with tensile strength which originate from at least one type of the class of inorganic polymers on a basis of silicates, preferably glass.
10. Product as claimed in claim 8, characterized by other fibres with tensile strength consisting of glass fibres, chopped strands with a length of 4-5 mm and a diameter of



0.013 mm and a ratio of length to diameter in the range of 300-400.

11. Product as claimed in claim 8,  
characterized by

5 other fibres with tensile strength of one or more types of the class of the natural biopolymers on a basis of cellulose, preferably from flax, jute, hemp, sisal, coconut, bamboo and miscanthus, wherein the percentage applied also depends on the number of external appendages on the fibres.

10 12. Product as claimed in claim 6,  
characterized in that

the polymer material consists of polypropylene, polystyrene, polyethylene or polyacrylate.

15 13. Product as claimed in claim 12,  
characterized in that

the polymer material consists of one or more olefinic homo- or copolymerisates with an M.F.I. (230/2,16) of 1-30 dg/min; preferably 2-15 dg/min; and which polymeric matrix material forms 10-50% by mass and preferably 15-40% by mass of the technical  
20 wood.

14. Product as claimed in any of the foregoing claims,  
characterized by

at least one additive for obtaining desired properties, which additive is added to the process flow at a suitable position  
25 in the compushtruding process during manufacture of the product.

15. Product as claimed in claim 14,  
characterized by

an additive with a desired influence on chosen properties of the product and belonging to at least one of the following  
30 classes:

- . influencers of adhesion between particles with tensile strength and matrix polymer (class H),
- . influencers of the properties of the surface of the product, particularly in respect of coatings or adhesives  
35 for applying in sandwich structures (class O),
- . influencers of the pyrogenic properties (class P),
- . influencers of the particle durability (class D),

- . blowing means for obtaining a foamed structure (class B),  
in the case of an unintended, sufficiently great  
temperature increase.

16. Product as claimed in claim 14,

5

characterized in that

as additives with a positive influence on the desired  
properties of the technical wood preferably one or more of the  
following are used, therein indicating the class or classes and  
the applied percentage related to the mass of technical wood:

10

	<u>Additive</u>	<u>Class</u>	<u>Mass-percentage</u>
	Polybond (Spider)	H O	1-3, preferably 2.5
	Polyweb (DOW)	H O	1-3, preferably 2.5
15	Exxelor (Exxon)	H O	1-3, preferably 2.5
	tributoxyethylphosphate	H O P	1-5, preferably 2
	1,2,3-propane triol	H O V	2-3, preferably 2.5
	diammoniumphosphate	P D	2-8
	ammoniumcarbonate	P B	1-3
20	ammoniumhydrogencarbonate	P B	
	1,2-ethane diol	O D	up to 3
	ethylacetate/ethanol 1/2 -1/3	O	up to 3
	methylacetate	H O	up to 4
	n-propanol	O	up to 1.5
25	silane A 171, 172, 174	H O D	0.5 - 3
	polyvinylacetate	H O D	up to 5
	surfactants, ionogenic/aniogenic, standard	H O	up to 2
	chlorinated polyolefins	H O	up to 5
30	UV-stabilizers	D	up to 3
	azodicarbonamide	P B	up to 3

17. Product as claimed in any of the foregoing  
claims,

35

characterized in that

at least one colouring agent or pigment is added to  
the product during manufacture.

18. Product as claimed in any of the foregoing  
claims,

characterized in that

the product consists substantially of a laminate comprising:

a plastic-based composite layer as claimed in any of  
5 the foregoing claims;

a first skin layer adhered thereto on one side and possessing chosen properties;

optionally a second skin layer adhered thereto on the other side and possessing chosen properties; and  
10 which layers are mutually adhered by for instance glueing, welding, mirror-welding, with an infrared laser, with hot air or other suitable treatment.

19. Product as claimed in claim 18,

characterized in that

15 the skin layer or at least one of the skin layers is of the type as claimed in any of the foregoing claims and the product principal direction thereof has a chosen direction relative to this product principal direction of the first mentioned plastic-based composite layer.

20 20. Apparatus for manufacturing a product as claimed in any of the foregoing claims, comprising:

a compounder operating at low pressure for plasticizing a mixture consisting substantially of plastic and particles with tensile strength, which  
25 particles are mixed with the plastic either beforehand or in the compounder, and pressing the plastic mixture to the outside via an outlet;

a rotating displacement pump further transporting the plastic mixture;

30 a distribution head further guiding the mixture in order to further transport the plastic mixture substantially as a plug flow;

orienting means further guiding the mixture, comprising at least one bundle of substantially parallel  
35 channels through which the plastic mixture can flow, which channels are dimensioned relative to the long particles such that, other than to a dominating extent in

the particle principal direction, they are too small to allow passage of the long particles present in the plastic mixture; and

5 a substantially prismatic mould head which connects onto the outlets of the channels and the form of which corresponds with the desired cross sectional form of the product;

such that the particle principal direction corresponds with the longitudinal direction of the mould head and the product principal direction;

10 which mould head is so long and has a temperature curve in the longitudinal direction such that at the end of the mouth the product has cooled to below its Vicat softening temperature.

15 21. Apparatus as claimed in claim 20, wherein the linear transverse dimensions of at least the parts of the channels located furthest downstream amount to a minimum of 2x the length of the short fibres and a maximum of 1.5x the length of the long fibres.

20 22. Apparatus as claimed in claim 20, comprising at least two bundles of channels mutually connecting in series, wherein the transverse dimensions of the channels of a bundle located further downstream are smaller than those of the channels of a bundle located further upstream.

25 23. Apparatus as claimed in claim 20, wherein the effective passages of the channels of the distribution head are adjustable, for instance by means of screws controllable from outside, or by means of selective heating.

30 24. Apparatus as claimed in claim 20, wherein at least one feed for adding particle material and/or additives to the plastic connects to the compounder.

35 25. Apparatus as claimed in any of the claims 20-24, comprising particle supply means which are adapted to introduce the particles into the compounder under a pressure such that these are compressed while air is expelled.

26. Apparatus as claimed in claim 25, comprising compression means for compressing the particle material to expel gases such as air prior to addition of the plastic.

5        27. Apparatus as claimed in any of the claims 20-26, wherein the compounder has a screw with a geometry such that the plastic mixture in which the particles are incorporated is successively compressed, decompressed and degassed under vacuum such that air and other gases are  
10 expelled from the mixture and from the pores present in the particles.

28. Method for manufacturing a product as claimed in any of the claims 1-17,

characterized in that

15        the finished product is subjected to an after-treatment comprising of heating the product to a temperature above the Vicat softening temperature of the applied plastic respectively the highest Vicat softening temperature of all the applied plastics, modelling the  
20 product to a desired shape and while maintaining a chosen pressure causing the product to cool in this shape to below the said Vicat softening temperature.

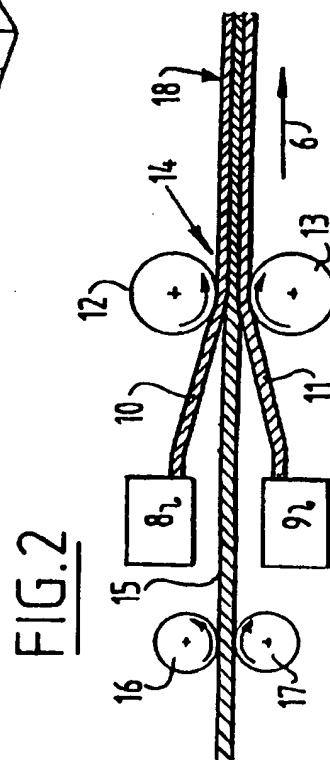
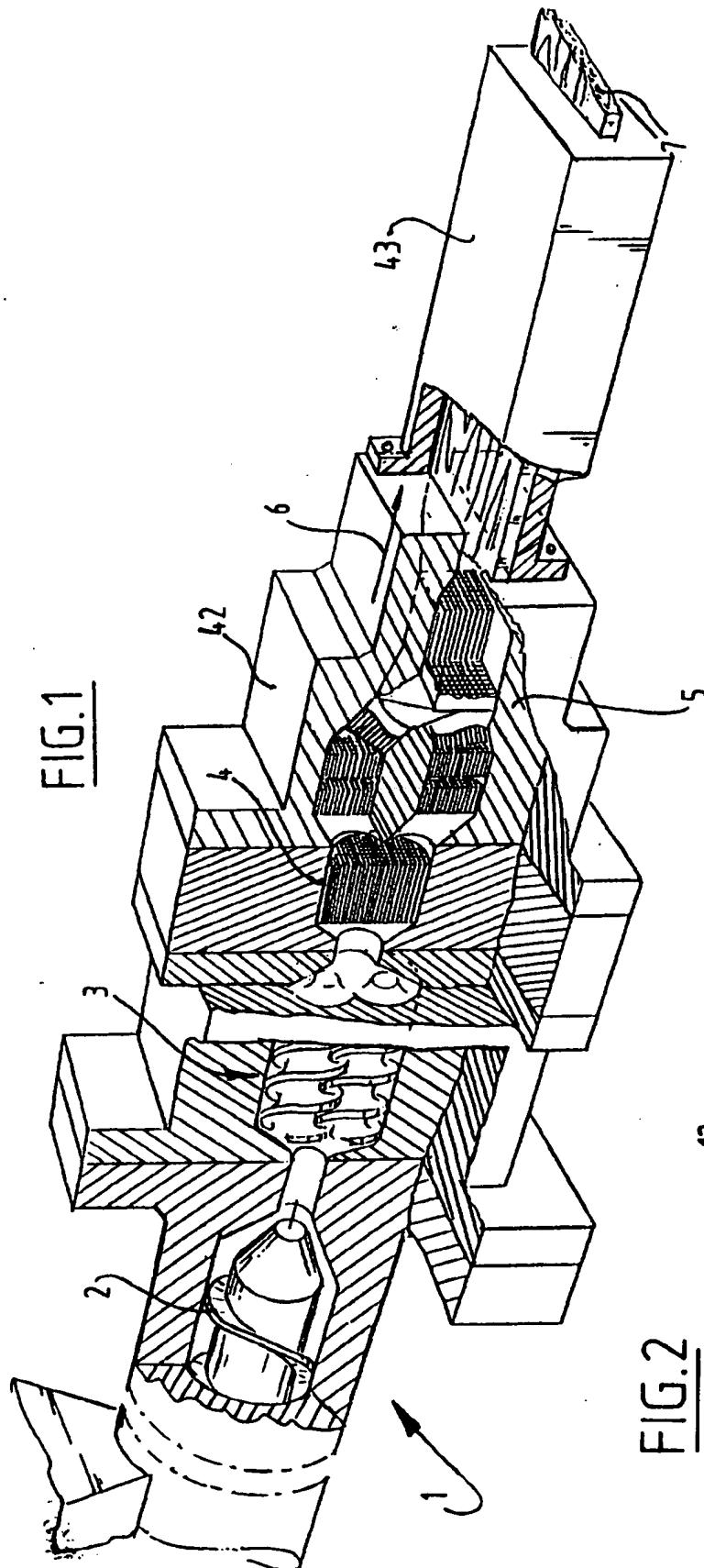
29. Product obtained with the method as claimed in claim 28.

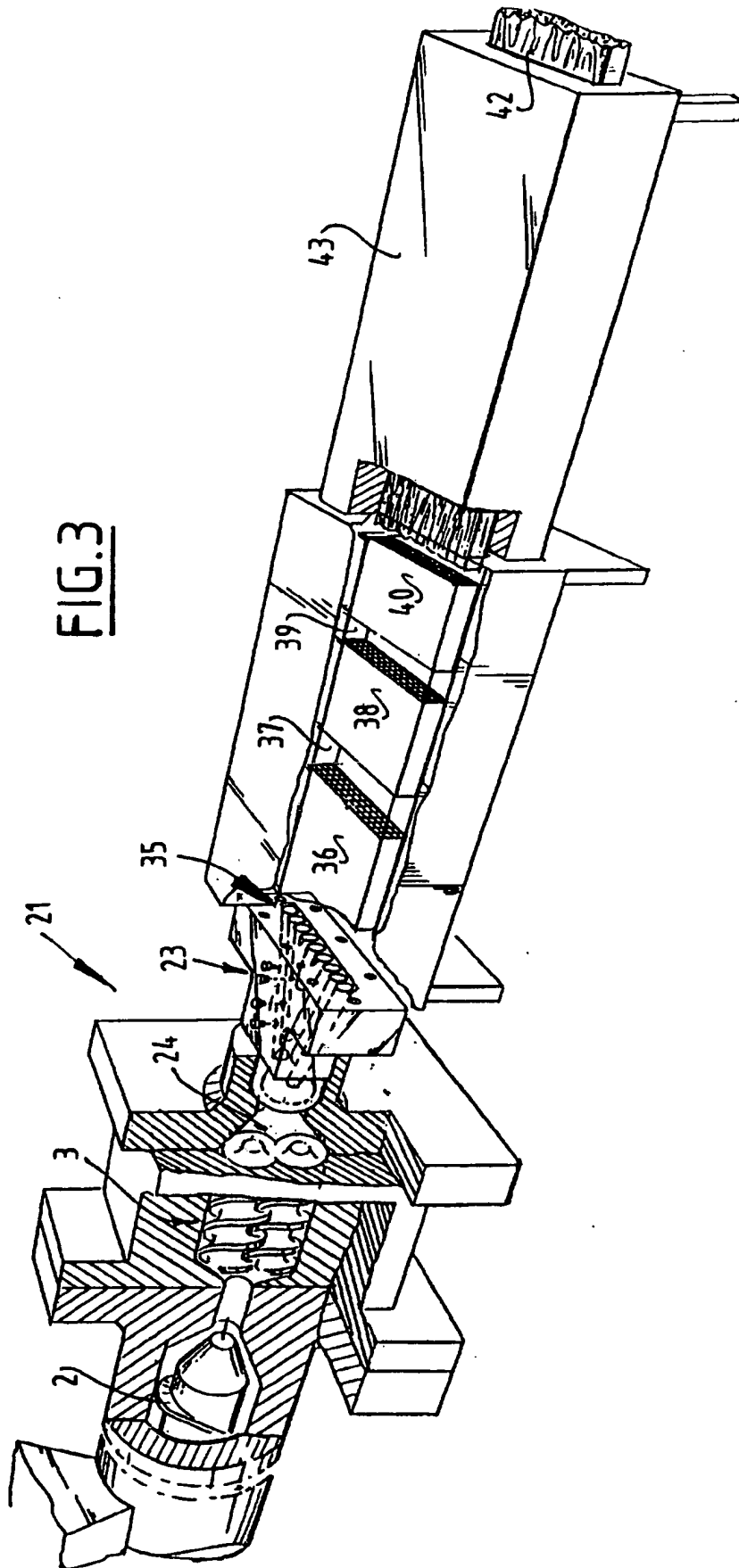
25        30. Method for manufacturing a product as claimed in claim 12,

characterized in that

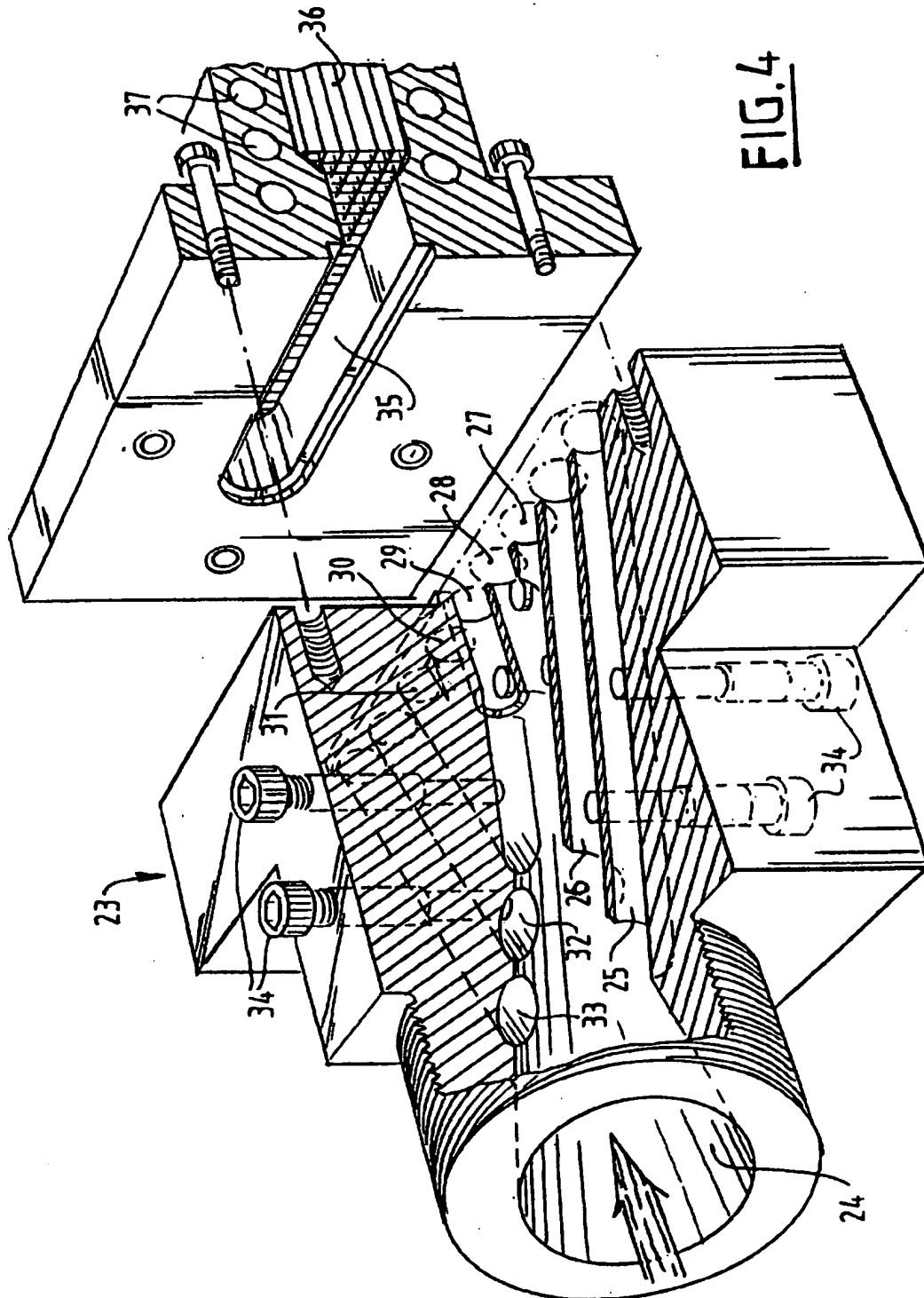
as starting polymer material is used a prepolymer on the basis of a styrene or an acrylate.

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# INTERNATIONAL SEARCH REPORT

Internat Application No  
PCT/NL 95/00153

A. CLASSIFICATION OF SUBJECT MATTER  
IPC 6 C08K7/00 C08J5/04 B29C47/00 B32B27/04 C08L101/00  
B29C71/02 C08F2/44 B29C67/24

According to International Patent Classification (IPC) or to both national classification and IPC

## B. FIELDS SEARCHED

Minimum documentation searched (classification system followed by classification symbols)  
IPC 6 C08K B29C

Documentation searched other than minimum documentation to the extent that such documents are included in the fields searched

Electronic data base consulted during the international search (name of data base and, where practical, search terms used)

## C. DOCUMENTS CONSIDERED TO BE RELEVANT

Category *	Citation of document, with indication, where appropriate, of the relevant passages	Relevant to claim No.
A	EP,A,0 319 589 (NAMBA) 14 June 1989	1,6,12
A	see page 7, line 1 - line 16; claims 1-12	1-13
	---	
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	see page 3, line 31 - page 4, line 17	
A	see claims 1,10,11,14	1,5
	---	
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	cited in the application	
	see column 4, line 21 - line 35	
	see column 7, line 7 - line 24; claims 1-7,10-15	
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☒ Further documents are listed in the continuation of box C.

☒ Patent family members are listed in annex.

### \* Special categories of cited documents :

- \*A\* document defining the general state of the art which is not considered to be of particular relevance
- \*E\* earlier document but published on or after the international filing date
- \*L\* document which may throw doubts on priority claim(s) or which is cited to establish the publication date of another citation or other special reason (as specified)
- \*O\* document referring to an oral disclosure, use, exhibition or other means
- \*P\* document published prior to the international filing date but later than the priority date claimed

- \*T\* later document published after the international filing date or priority date and not in conflict with the application but cited to understand the principle or theory underlying the invention
- \*X\* document of particular relevance; the claimed invention cannot be considered novel or cannot be considered to involve an inventive step when the document is taken alone
- \*Y\* document of particular relevance; the claimed invention cannot be considered to involve an inventive step when the document is combined with one or more other such documents, such combination being obvious to a person skilled in the art.
- \*G\* document member of the same patent family

Date of the actual completion of the international search

29 March 1996

Date of mailing of the international search report

26.04.96

Name and mailing address of the ISA

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Engel, S

# INTERNATIONAL SEARCH REPORT

Internat. Application No  
PCT/NL 95/00153

C(Continuation) DOCUMENTS CONSIDERED TO BE RELEVANT		
Category *	Citation of document, with indication, where appropriate, of the relevant passages	Relevant to claim No.
A	DE,A,28 11 795 (POLYPLASTICS) 12 October 1976 see page 5, paragraph 2 see page 7, paragraph 2; claims 1-4; example 1 ---	1,5
A	DATABASE WPI Section Ch, Week 8504 Derwent Publications Ltd., London, GB; Class A17, AN 85-024748 & SU,A,1 100 288 ( ZLOBINA V A ) , 30 June 1984 see abstract ---	1-13
A	DATABASE WPI Section Ch, Week 8504 Derwent Publications Ltd., London, GB; Class A17, AN 85-022209 & JP,A,59 217 743 ( KASAI KOGYO KK ) , 7 December 1984 see abstract ---	1-13
A	PATENT ABSTRACTS OF JAPAN vol. 94 no. 00010 & JP,A,62 079615 (CHISSO CORP) 4 October 1994, see abstract & DATABASE WPI Week 9444 Derwent Publications Ltd., London, GB; AN 94-354843 & JP,A,62 079 615 (CHISSO) , 4 October 1994 see abstract ---	1-19
E	NL,A,9 302 125 (AVIPLAST) 3 July 1995 see page 4, line 30 - page 5, line 2 see page 5, line 11 - line 28 see figures 1,2 see claims 1,11 & NL,A,9 400 308 (AVIPLAST) see page 3, line 23 - line 39 see page 5, line 13 - page 6, line 20; claims 1,4,5,8,10,11; figure 1 ---	20
E		1,12,20, 21
A	US,A,5 096 406 (BROOKS ET AL.) 17 March 1992 see figures & US,A,5 082 605 (BROOKS ET AL.) cited in the application see column 7, paragraph 2 ---	20,21
A		20
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# INTERNATIONAL SEARCH REPORT

Intern. Application No  
PCT/NL 95/00153

C.(Continuation) DOCUMENTS CONSIDERED TO BE RELEVANT		
Category *	Citation of document, with indication, where appropriate, of the relevant passages	Relevant to claim No.
A	PATENT ABSTRACTS OF JAPAN vol. 014 no. 305 (M-0992) ,29 June 1990 & JP,A,02 098429 (NHK SPRING CO LTD) 10 April 1990, see abstract ---	1,28,29
A	EP,A,0 008 143 (SOLVAY & CIE.) 20 February 1980 see page 3, line 33 - page 4, line 5 see page 6, line 9 - line 11 see page 8, line 8 - line 15 ---	1,28,29
A	US,A,5 401 154 (M.M. SARGENT) 28 March 1995 see column 3, line 19 - line 28 ---	1,28,29
A	DATABASE WPI Section Ch, Week 8522 Derwent Publications Ltd., London, GB; Class A32, AN 85-131721 & JP,A,60 069 161 ( TOSHIBA KK) , 19 April 1985 see abstract -----	1,30

# INTERNATIONAL SEARCH REPORT

International application No.

PCT/NL95/00153

## Box I Observations where certain claims were found unsearchable (Continuation of item 1 of first sheet)

This international search report has not been established in respect of certain claims under Article 17(2)(a) for the following reasons:

1. ☐ Claims Nos.:  
because they relate to subject matter not required to be searched by this Authority, namely:
2. ☐ Claims Nos.:  
because they relate to parts of the international application that do not comply with the prescribed requirements to such an extent that no meaningful international search can be carried out, specifically:
3. ☐ Claims Nos.:  
because they are dependent claims and are not drafted in accordance with the second and third sentences of Rule 6.4(a).

## Box II Observations where unity of invention is lacking (Continuation of item 2 of first sheet)

This International Searching Authority found multiple inventions in this international application, as follows:

- 1.- claims 1-19: Plastic composite, products and process
- 2.- claims 20-27: Apparatus and its technical features
- 3.- claims 28-29: Method, using after-treatment
- 3.- claim 30: method, starting from prepolymer

1. ☒ As all required additional search fees were timely paid by the applicant, this international search report covers all searchable claims.
2. ☐ As all searchable claims could be searched without effort justifying an additional fee, this Authority did not invite payment of any additional fee.
3. ☐ As only some of the required additional search fees were timely paid by the applicant, this international search report covers only those claims for which fees were paid, specifically claims Nos.:
4. ☐ No required additional search fees were timely paid by the applicant. Consequently, this international search report is restricted to the invention first mentioned in the claims; it is covered by claims Nos.:

Remark on Protest

☒ The additional search fees were accompanied by the applicant's protest.

☐ No protest accompanied the payment of additional search fees.

# INTERNATIONAL SEARCH REPORT

Information on patent family members

Internat. Application No  
PCT/NL 95/00153

Patent document cited in search report	Publication date	Patent family member(s)	Publication date
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US-A-5401154	28-03-95	NONE	